# Control of Surface Plasmon-Polaritons in Magnetoelectric Heterostructures

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Abstract—The paper is devoted to the investigation of the surface plasmon polaritons in metal-dielectric heterostructures containing materials with magnetoelectric properties. The analysis of various possible configurations shows that the magnetoelectric effect influences the plasmonic surface wave polarization, localization, and dispersion. The essential case of  $Cr_2O_3$  as a magnetoelectric dielectric is described in detail, revealing the switching between different regimes of magnetoelectric impact on plasmon-polaritons. Plasmonic heterostructures with magnetoelectric constituents can be used as a polarization- and dispersion-sensitive instrument to reveal the magnetoelectric coupling. Apart from that, the spinflop effect provides an efficient tool for control of the far-field optical response of plasmonic structures by the external magnetic field.

*Index Terms*—Antiferromagnetics, magnetoelectric effect, plasmonics.

### I. INTRODUCTION

**N** OWADAYS, control of surface plasmon polaritons (SPPs) by an external stimulus is of prime importance for telecommunication and other applications and is usually referred to as active plasmonics [1]–[4]. Active plasmonics utilizes strong dependence of the SPP properties, namely its dispersion, localization, the transverse profile and polarization on

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the permittivity of the surrounding media. Slight variations of the dielectric permittivity result in a significant shift of the SPP resonance which is widely used nowadays for sensing applications [5]–[8] and all-optical light control [9]–[14]. The impact of magneto-optical gyrotropy on the SPP properties is promising for ultrafast modulation of laser radiation [15]–[18], while the presense of optical activity [19] performs modification of the SPP polarization. It was shown that such issues of magnetoelectric coupling as chirality [20] and the axion effect in topological insulators [21] can be revealed in plasmonic structures via observation of the changes in the SPP excitation conditions or polarization mediated by them.

Modification of the SPP properties in the presense of the magnetoelectric (ME) effect is very promising in the context of the SPP control. Although surface electromagnetic waves at the interfaces of bianisotropic dielectrics have been studied earlier [22], [23], the properties of the SPP waves supported by metal/ME dielectric structures were not addressed before. At the same time, compared to the surface electromagnetic waves in dielectrics, SPPs are characterized by stronger electromagnetic field concentration in a region of about 100 nm and higher sensitivity to the medium properties which makes them promising for efficient ME light control.

In Section II of this paper we provide a theory of SPP waves in a most general case of a metal/ME dielectric interface. We provide the description and classification of different ME configurations and phases corresponding to different orientations and different types of ME crystals which makes it possible to predict the variations of SPP characteristics in the particular structure only by the form of its ME tensor. In Section III we apply our theory to  $Cr_2O_3$  crystal which not only has the ME response but also provides the possibility to switch it due to the spin-flop effect. We analytically and numerically investigate the SPP modes in several basic configurations. We show how laser radiation can be modulated in the far-field using SPPs in ME plasmonic structures. Although the effects found in Cr<sub>2</sub>O<sub>3</sub> are rather moderate, similar approach can be applied to other crystals exhibiting stronger ME response. For example, artificial metamaterials and composite multilayered structures like  $BaTiO_3/CoFe_2O_4$ or  $BaTiO_3/BiFeO_3$  are designed to provide the enhancement of the ME effects [24]–[27]. The structure that is characterized by the typical value of the ME constant of 0.1 has recently been designed [28], that makes the optical ME effects of practical interest.

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## II. THEORETICAL APPROACH TO THE MAGNETOELECTRIC SURFACE PLASMON POLARITONS

## A. ME Effect in $Cr_2O_3$ Crystal

The ME effect is a coupling between the electric and magnetic field components so that the magnetic field induces electric polarization and the electric field induces magnetization in a material. It can be observed in antiferromagnetics, multiferroics, and topological insulators [29]–[32] as well as in some artificial metamaterials [24]. The following constitutive equations describe the ME effect at optical frequencies (permeability is set to unity) [26]:

$$\mathbf{D} = \hat{\varepsilon}^0 \mathbf{E} + \hat{\alpha} \mathbf{B}, \ \mathbf{H} = \mathbf{B} - \hat{\alpha}^+ \mathbf{E}$$
(1)

where  $^+$  is complex conjugation, and  $\hat{\alpha}$  is the ME susceptibility tensor whose symmetry and characteristic component values depend on the material. Energy conservation law requires the ME susceptibility tensor  $\hat{\alpha}$  to be the same in both of equations (1) for all of the non-absorbing materials. We restrict our consideration to the case of real components of  $\hat{\alpha}$ .

The case of the isotropic form of the ME susceptibility tensor is similar to the so-called axion effect [33]–[35] which may appear if a perturbation breaking time reversal symmetry is present. The non-diagonal ME tensor corresponds to the toroidal type of ME response that is related to the non-zero toroidal moment and has fundamental interest for plasmonic structures [36], [37].

One of the prominent representatives of the ME materials is chromium (III) oxide  $Cr_2O_3$  [38]. Its symmetry leads to the following form of the ME susceptibility tensor [39]:

$$\hat{\alpha} = \begin{pmatrix} \lambda_1 L_{y'} + \lambda_3 L_{z'} & \lambda_1 L_{x'} & \lambda_2 L_{x'} \\ \lambda_1 L_{x'} & -\lambda_1 L_{y'} + \lambda_3 L_{z'} & \lambda_2 L_{y'} \\ \lambda_4 L_{x'} & \lambda_4 L_{y'} & \lambda_5 L_{z'} \end{pmatrix}$$
(2)

where **L** is the unitary antiferromagnetic vector and  $\lambda_j$ s are the coefficients that for the selected material have the typical values of the order of  $10^{-4}$  [40]. This ME susceptibility tensor is written in the local coordinate system associated with the crystal symmetry: Ox' axis is parallel to the two fold axis and Oz' axis is parallel to the three fold symmetry axis (the *c*-axis) [41].

An important feature of  $Cr_2O_3$  is that its ME response can be switched between two types below the Neel temperature of  $T_N$ = 307 K. Such switching is possible due to the "spin-flop" effect that is a reorientation of the spins in strong magnetic field applied along the three-fold axis and exceeding the threshold value of about 7 T [42]. Below the threshold, in so-called low-field phase, the spins and the antiferromagnetic vector of  $Cr_2O_3$  both are oriented along the *c*-axis. At this the ME susceptibility tensor has the diagonal form, so that the electrodynamics demonstrates the axion-like behaviour. Spin-flop results in the 90° rotation of the antiferromagnetic vector that becomes perpendicular to the *c*-axis [40], [43]. The ME tensor in this case has non-diagonal components that affect the electrodynamics of the crystal in a different way compared to the low-field phase.

Therefore, the spin-flop is a transition from axion-like to the toroidal magnetoelectricity. Due to that,  $Cr_2O_3$  is a unique object on which one can trace the similarities and differences between the axion-like and toroidal effects. That makes  $Cr_2O_3$ an appropriate model object for the study of fundamental properties of ME SPPs. Due to the switchable ME effect,  $Cr_2O_3$ is perspective for active plasmonics. Moreover, although the changes in the ME susceptibility tensor produced by spin-flop transition are rather small, it might be possible to reveal both the ME effect and the spin-flop phase transition by the sensitive SPP resonance. Therefore, the plasmonic surface of a  $Cr_2O_3$ crystal coated with a metal, e.g., gold, provides an optical tool for investigation of the  $Cr_2O_3$  phase transitions and allows for a light control via applied external static magnetic field.

#### B. General Method of the ME SPPs Analysis

For the analysis of ME SPP properties we find the eigenmodes of Maxwell's equations with the corresponding boundary conditions and constitutive equations (1) [44]. In the absence of the surface charge and the surface current the boundary conditions take the conventional forms with normal components of  $\mathbf{D}$ and  $\mathbf{B}$ , as well as the tangential components of  $\mathbf{E}$  and  $\mathbf{H}$  being continuous over the interface.

The ME susceptibility tensor occuring in constitutive equations (1) can be decomposed to symmetric and antisymmetric parts:  $\hat{\alpha} = \hat{\alpha}^S + \hat{\alpha}^{AS}$ . Antisymmetric part is associated with the toroidal moment [39], [45] and can be expressed as:

$$\alpha_{ij}^{AS} = \epsilon_{ikj} \tau_k, \tag{3}$$

where  $\epsilon_{ijk}$  is Levi-Civita symbol, and characteristic vector  $\boldsymbol{\tau}$  is proportional to the toroidal moment vector  $\mathbf{T}$  that is dual to  $\hat{\alpha}^{AS}$ .

For the most general description, the electromagnetic field of the SPP eigenmode inside of a ME dielectric is represented as the sum of the two partial TE and TM components with different polarization ( $\mathbf{E}_j$ ) and localization ( $\gamma_j$ ), in general, but equal propagation constants  $\beta$ :

$$\mathbf{E}_{\rm spp}(z>0) = \left(\mathbf{E}_{\rm TM} e^{-\gamma_{\rm TM} k_0 z} + \mathbf{E}_{\rm TE} e^{-\gamma_{\rm TE} k_0 z}\right) e^{i\beta k_0 x}$$
(4)

while in the metal field has the conventional form

$$\mathbf{E}_{\rm spp}(z<0) = \mathbf{E}_m e^{\gamma_m \, k_0 \, z} e^{i\beta k_0 \, x} \tag{5}$$

The localization coefficients and the propagation constant are normalized by vacuum wavenumber  $k_0$ . Detailed information on the ME-induced  $\gamma_j$  variation can be found in Supplementary. The coordinate system is chosen as shown in Fig. 1(a). For the convenience, we denote interface plane as  $\Xi$ , and SPP propagation direction as  $\beta$ .

Using the above plane-wave representation for the SPP eigenmode with complex wave vector  $\kappa^{(j)} = \{\beta; 0; i\gamma_j\}$ , Maxwell's equations and the constitutive equations (1) and (3) one can obtain the following wave equation:

$$[\boldsymbol{\kappa} \times [\boldsymbol{\kappa} \times \mathbf{E}]] + \hat{\varepsilon}(\boldsymbol{\kappa})\mathbf{E} = 0$$
(6)

where the effective dielectric permittivity tensor  $\hat{\varepsilon}(\kappa)$  that includes ME contribution has the form

$$\varepsilon_{ij}(\boldsymbol{\kappa}) = \varepsilon_{ij}^0 - (\kappa_s(\epsilon_{isk}\alpha_{kj}^S - \epsilon_{ksj}\alpha_{ik}^S) + \kappa_j\tau_i + \kappa_i\tau_j - 2(\boldsymbol{\kappa}\boldsymbol{\tau})\delta_{ij})$$
(7)

where  $\delta_{ij}$  is Kronecker delta.



Fig. 1. (a) Possible configurations of the SPP propagation at the metal/ $Cr_2O_3$ interface. (b) The birefringence of the SPP modes at  $Cr_2O_3$ –gold (black solid line) or silver (red solid line) interfaces. Vertical dashed lines correspond to surface plasmon frequencies  $\omega_{sp}$  while gray dash-dot line shows the birefrigence asymptote. Optical parameters of metals are given in Supplementary.

The algebraic equation (6) is rather complicated, and to get clear analytical results, we apply the linear in  $\hat{\alpha}$  approximation. That means, since normally  $|\alpha_{ij}| << \varepsilon_{mn}^0$ , we can neglect all terms that are associated with powers of  $\alpha$  higher than first, and find the eigenmodes of (6) in the form (4), (5). Thus we obtain all the information about the ME SPP properties: polarization, localization, dispersion in the explicit form. This method is used for the analysis of the ME SPPs in all of the considered further in the paper configurations.

However in many real cases the analytical soltuion of (6) is complicated, especially in the case of the considered  $Cr_2O_3$  that has anisotropic permittivity tensor  $\hat{\varepsilon}^0$  and both symmetric and antisymmetric components of the ME susceptibility tensor. In order to reveal the impact of the symmetric and antisymmetric parts of the ME susceptibility tensor on the SPP separately, we first address the idealized problems of isotropic media with fully symmetric or fully antisymmetric ME tensors. For the simplicity, we restrict the former case to the case of isotropic ME tensor that corresponds to the so-called axion coupling.

## C. Axion-Type Impact on SPPs

The impact of the axion effect on the electromagnetic wave properties reveals itself at the interface between axion and nonaxion medium while the bulk electromagnetic wave does not experience the influence of the axion effect. The peculiarities of the transmission or reflection of bulk electromagnetic waves in structures with axion/non-axion layers were considered earlier [46]–[48]. We focus our attention on the SPP properties at axion-type ME/metal interface.

Here we consider an isotropic dielectric possessing the dielectric permittivity  $\varepsilon^0$  with a diagonal ME susceptibility tensor in the form  $\hat{\alpha} = \alpha \hat{I}$ , where  $\hat{I}$  is the identity tensor.

The analysis of the SPP properties shows that the dispersion has no linear-in- $\alpha$  terms so the axion effect does not noticeably change the propagation constant and the SPP resonance frequency. Since there is a polarization degeneracy of the bulk modes in a medium with the axion effect, partial TM and TE components of the SPP wave should have equal localization coefficients  $\gamma_{\rm TM} = \gamma_{\rm TE}$ . Notice that due to the axion coupling between the TM and TE components they are both present in the SPP field. The analytical solution of the SPP eigenmode problem (taking  $H_y = 1$  here and elsewhere) gives the following expressions for the field in axion media:

$$\mathbf{E} = \begin{pmatrix} i\frac{\gamma_{\mathrm{TM}}}{\varepsilon^{0}} \\ -\alpha\frac{1}{\varepsilon^{0} - \varepsilon_{m}} \\ -\frac{\beta}{\varepsilon^{0}} \end{pmatrix}, \quad \mathbf{H} = \begin{pmatrix} i\alpha\frac{\gamma_{\mathrm{TM}}\varepsilon_{m}}{\varepsilon^{0}(\varepsilon^{0} - \varepsilon_{m})} \\ 1 \\ -\alpha\beta\frac{\varepsilon_{m}}{\varepsilon^{0}(\varepsilon^{0} - \varepsilon_{m})} \end{pmatrix}. \quad (8)$$

Let us define the ratio between TE and TM components as  $E_u/H_u$  that in the case of the axion-type ME effect is  $E_y/H_y = -\frac{\alpha}{\varepsilon^0 - \varepsilon_m}$ . The SPP polarization ellipse in non-ME case was oriented in Oxz plane. Due to the axion-type ME effect polarization ellipse experiences the slight turn around Oxaxis by an angle of  $\varphi = E_y/E_z$  (since  $E_y$  and  $E_z$  are in-phase). It is important that the polarization of the SPP is constant during its propagation along the metal-dielectric interface. Similar effect was reported earlier for gyrotropic interfaces, in contrast to Faraday rotation or optical activity-caused rotation of the bulk waves. While the ME susceptibility tensor is isotropic, the direction of this polarization turn is strictly defined by the SPP propagation direction, since the angle is odd in  $\beta$ . This may be referred as the locking between the SPP propagation direction and the orientation of the polarization ellipse, similarly to the spin-momentum locking for electronic surface states.

Similar analysis of the SPPs at the plasmonic interface with isotropic axion-type ME effect can be directly applied to the topological insulators [21] with  $\alpha = 1/137$  that possess the axion properties due to the perturbations of the time-reversal symmetry [34], [35].

#### D. Impact of Toroidal Magnetic Moment on SPPs

The presence of the toroidal moment leads to the effect of non-reciprocal wave propagation, so that the waves propagating along  $\tau$  and opposite to  $\tau$  possess different wavenumbers [49]. The relations between **E** and **H** vectors are also modified and, moreover, **E** and **H** vectors are generally no longer perpendicular to the wavevector. The essential point is that the impact of the toroidal moment on the bulk wave properties is polarizationindependent [49], similarly to the case of the axion effect.

The properties of SPPs in media with toroidal moment are strongly dependent on the direction of  $\tau$ . There are three basic configurations:

- 1) the longitudinal configuration:  $\tau \parallel \Xi, \tau \parallel \beta (\tau \parallel Ox);$
- 2) the transversal configuration:  $\tau \parallel \Xi$ ,  $\tau \perp \beta (\tau \parallel Oy)$ ;
- 3) the polar configuration:  $\tau \perp \Xi \ (\tau \parallel Oz)$ .

In the longitudinal configuration propagation constant and localization coefficients are affected by the toroidal moment

$$\beta = \sqrt{\frac{\varepsilon_m \varepsilon^0}{\varepsilon_m + \varepsilon^0}} - \tau_x \frac{\varepsilon_m^2}{\varepsilon_m^2 - (\varepsilon^0)^2}.$$
 (9)

Equation (9) demonstrates the nonreciprocal propagation of the SPP: the SPPs propagating along  $\tau$  and opposite to  $\tau$  possess different propagation constants. The polarization of the SPP remains pure TM. However, the relations between the field

 TABLE I

 The ME Impact on the SPP Modes in Idealized Cases

Configuration		SPP characteristics variation	
The axion effect		Polarization (ellipse is tilted around $Ox$ axis, $E_y/H_y$ is real)	
The toroidal	$egin{array}{c}  au \parallel \Xi, \ eta \parallel  au \end{array} \ eta \parallel  au \end{array}$	Dispersion (non-reciprocal), localization in both media, the eccentricity of the polarization ellipse	
effect	$egin{array}{c}  au \parallel \Xi, \ eta \perp  au \end{array}$	Polarization (ellipse is tilted around $Oz$ axis, $E_y/H_y$ is imag.)	
	$ au \perp \Xi$	Localization depth in dielectric	

components inside the dielectric are slightly modified

$$\mathbf{E} = \begin{pmatrix} i \frac{1}{\sqrt{-(\varepsilon_m + \varepsilon^0)}} - i\tau_x \frac{\sqrt{-\varepsilon_m \varepsilon^0}}{\varepsilon_m^2 - (\varepsilon^0)^2} \\ 0 \\ -\sqrt{\frac{\varepsilon_m}{\varepsilon^0(\varepsilon_m + \varepsilon^0)}} + \tau_x \frac{\varepsilon^0}{\varepsilon_m^2 - (\varepsilon^0)^2} \end{pmatrix}, \ \mathbf{H} = \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}.$$
(10)

It follows from (10) that if the SPP propagates along  $\tau$  the electric field components diminish and, moreover, the polarization ellipse becomes narrower as its semiaxes ratio  $E_x/E_z$  gets smaller. At the same time, the semiaxes ratio of the polarization ellipse for the metal  $E_z/E_x$  also diminishes. For the opposite direction of the SPP propagation the results are opposite.

In the polar configuration only localization constant for the dielectric changes upon the toroidal moment. Since the configuration is isotropic in the interface plane, the SPP propagation is reciprocal and the field components remain unchanged.

On the contrary, in the transversal configuration the propagation constant and the localization coefficients are independent of the toroidal moment. At the same time, the SPP polarization acquires TE components. Due to the optical isotropy  $\gamma_{\rm TM} = \gamma_{\rm TE}$ . The field inside the dielectric has a form

$$\mathbf{E} = \frac{1}{\varepsilon^0} \begin{pmatrix} i\gamma_{\mathrm{TM}} \\ i\tau_y \frac{\beta}{\gamma_{\mathrm{TM}} + \gamma_m} \\ -\beta \end{pmatrix}, \ \mathbf{H} = \begin{pmatrix} -\tau_y \frac{\gamma_m \beta}{\varepsilon^0 (\gamma_{\mathrm{TM}} + \gamma_m)} \\ 1 \\ i\tau_y \frac{\varepsilon^0 - \gamma_{\mathrm{TM}} \gamma_m}{\varepsilon^0 (\gamma_{\mathrm{TM}} + \gamma_m)} \end{pmatrix}$$
(11)

so the TE/TM ratio is  $E_y/H_y = i\tau_y \sqrt{-\frac{\varepsilon_m}{\varepsilon^0}} \frac{1}{\varepsilon^{0}-\varepsilon_m}$ .

The plane of the polarization ellipse turns around Oz axis by the angle of  $\varphi = E_y/E_x$  compared to the non-toroidal case (unlike the case of the axion effect,  $E_x$  and  $E_y$  are in-phase), and upon the toroidal moment reversal the polarization ellipse turns in the other direction.

The results of Sections II-C and II-D are summarized in Table I.

## III. SURFACE PLASMON POLARITONS AT THE METAL/ $Cr_2O_3$ INTERFACE: OPTICAL ANISOTROPY AND MAGNETOELECTRICITY

## A. General Features of the SPP at the Metal/Cr<sub>2</sub>O<sub>3</sub> Interface

The observed impact of the ME effect in a  $Cr_2O_3$  crystal on the SPP modes depends significantly on the relative orientation of the crystal *c*-axis and the antiferromagnetic vector  $\mathbf{L}$  with respect to the plasmonic interface  $\Xi$  between a metal and the Cr<sub>2</sub>O<sub>3</sub> crystal as well as with respect to the direction of the SPP propagation  $\beta$ . For the detailed analysis we choose 9 basic possible configurations of the SPP propagation along the interface [see Fig. 1(a)] which correspond to 3 possible orientations of the *c*-axis and 3 possible orientations of  $\mathbf{L}$  vector with respect to  $\beta$  direction.

 $Cr_2O_3$  crystal possesses optical anisotropy, for the wavelength of 633 nm  $\varepsilon_{\perp} = 4.9$  and  $\varepsilon_{\parallel} = 5.15$  [41] where  $\varepsilon_{\perp}$  and  $\varepsilon_{\parallel}$  are components of  $\hat{\varepsilon}^0$  corresponding to the perpendicular or parallel to the *c*-axis directions. Therefore the dispersion of the SPP waves at its interface depends on crystal orientation. In the case of *c*-axis  $\parallel \Xi, \beta \perp c$ -axis, the SPP can be called "ordinary" since it does not experience anisotropy:

$$\beta_o = \sqrt{\frac{\varepsilon_\perp \varepsilon_m}{\varepsilon_\perp + \varepsilon_m}}.$$
 (12)

In the other cases optical anisotropy affects the SPP and its "extraordinary" dispersion takes the following forms:

$$\beta_e = \sqrt{\frac{\varepsilon_m \varepsilon_{\parallel} (\varepsilon_m - \varepsilon_{\perp})}{\varepsilon_m^2 - \varepsilon_{\perp} \varepsilon_{\parallel}}}$$
(13)

if the c-axis is oriented perpendicular to the interface  $\Xi$ ; and

$$\beta_e = \sqrt{\frac{\varepsilon_m \varepsilon_\perp (\varepsilon_m - \varepsilon_\parallel)}{\varepsilon_m^2 - \varepsilon_\perp \varepsilon_\parallel}}$$
(14)

if  $\beta \parallel c$ -axis. Such dependence of the dispersion on the orientation of the anisotropic crystal was presented in [50].

Numerical estimations of the SPP mode "birefringence" for the two perpendicular  $\beta$  directions in the case of *c*-axis  $\parallel \Xi$  [see Fig. 1(b)] show that the difference between the wavenumbers of the two modes increases near the surface plasmon frequency  $\omega_{sp}$ at which  $\beta(\omega_{sp}) = \infty$ . There is a slight difference between the surface plasmon frequencies of the two modes according to (12) and (14). For example, for the gold/Cr<sub>2</sub>O<sub>3</sub> interface the surface plasmon frequency corresponds to  $\lambda_{sp e} = 506$  nm and  $\lambda_{sp o} =$ 503.5 nm while for the silver/Cr<sub>2</sub>O<sub>3</sub> interface  $\lambda_{sp e} = 390$  nm and  $\lambda_{sp o} = 387$  nm. For higher wavelengths ( $\lambda > 900$  nm for the considered structure) the birefrigence reaches its asymptote ( $\beta_e - \beta_o$ )/ $\beta_o = \sqrt{\varepsilon_{\parallel}/\varepsilon_{\perp}} - 1$  and equals 2.6% in Cr<sub>2</sub>O<sub>3</sub>.

The analysis of the SPP mode characteristics in  $Cr_2O_3$ /metal structure with the corresponding ME response was performed as described in Section II-B. Table II summarizes its results showing which changes in mode properties corresponding to the certain configurations and the crystal phases are observed. The analytical expressions for the corresponding changes are rather complicated nevertheless Table II gives an overview of the nature of the ME impact on the SPP modes. Further we discuss several configurations in a more detailed way and perform numerical simulations using the following values for the ME coefficients:  $\lambda_1 = 1.6 \cdot 10^{-4}$ ,  $\lambda_2 = 1.0 \cdot 10^{-4}$ ,  $\lambda_3 = -1.965 \cdot 10^{-4}$ ,  $\lambda_5 = -0.82 \cdot 10^{-4}$  [41].

TABLE II CHARACTERISTICS OF SPP MODES THAT ARE AFFECTED BY THE ME EFFECT IN  $Cr_2O_3$ /Metal Structure for Various Configurations and Phases

$c \perp$ interface						
The low-field	The spin-flop phase ( $\mathbf{L} \perp c$ )					
phase ( $\mathbf{L} \parallel c$ )	$SPP\perp \mathbf{L}$		SPP $\parallel \mathbf{L}$			
polarization $(\frac{E_y}{H_y}$ is real)	dispe locali:	dispersion localization				
c    interface						
The low-field	The spin-flop phase ( $\mathbf{L} \perp c$ )					
phase ( $\mathbf{L} \parallel c$ ), any	$\mathbf{L} \perp$ interface		L    interface, any			
SPP direction	$SPP \parallel c$	$\mathrm{SPP}\perp c$	SPP direction			
polarization $(\frac{E_y}{H_y}$ is real)	polarization $(\frac{E_y}{H_y}$ is imag.) localization in dielectric	dispersion localization	localization in dielectric			

## B. SPP Polarization in the Low-Field Phase of $Cr_2O_3$

Let us discuss ME SPP properties in the low-field phase of  $\operatorname{Cr}_2O_3$  (which means  $\mathbf{L} \parallel c$ -axis). This case corresponds to the axion effect described in Section II-C, therefore one should expect change only in SPP polarization. In both possible basic low-field configurations:  $\mathbf{L} \parallel c$ -axis  $\parallel \Xi$  and  $\mathbf{L} \parallel c$ -axis  $\perp \Xi$ , the SPP mode has both nonzero TM and TE components with different localization coefficients that are independent of magnetoelectric susceptibility tensor components. The TE/TM ratio for the "extraordinary" SPP (for the case *c*-axis  $\perp \Xi$ ) inside  $\operatorname{Cr}_2O_3$  is

$$\frac{E_y}{H_y} = \frac{\lambda_5 - \lambda_3}{\varepsilon_\perp - \varepsilon_\parallel} - \left[ (\gamma_m + \gamma_{\rm TM}) \frac{\lambda_5 - \lambda_3}{\varepsilon_\perp - \varepsilon_\parallel} + \gamma_{\rm TM} \frac{\lambda_5}{\varepsilon_\perp} \right] \frac{e^{-(\gamma_{\rm TE} - \gamma_{\rm TM})|z|}}{\gamma_m + \gamma_{\rm TE}}$$
(15)

The analogous expression can be obtained for the "ordinary" SPP

$$\frac{E_y}{H_y} = \frac{\lambda_5 - \lambda_3}{\varepsilon_\perp - \varepsilon_\parallel} - \left[ \frac{\lambda_5 - \lambda_3}{\varepsilon_\perp - \varepsilon_\parallel} + \frac{\lambda_3 \gamma_{\rm TM}}{\varepsilon_\perp (\gamma_{\rm TE} + \gamma_m)} \right] e^{-(\gamma_{\rm TE} - \gamma_{\rm TM})|z|} \quad (16)$$

The polarization ellipse turns around Ox axis (that coincides with  $\beta$ ) with respect to the non-ME case, see Fig. 2(a) ( $E_y$  and  $E_z$  are in-phase). At the same time according to (15), (16) the polarization of the SPP depends on z inside Cr<sub>2</sub>O<sub>3</sub>. According to the simulations presented in Fig. 2(c) and (d) in both cases the TE/TM ratio increases with the distance to the interface and at  $k_0 z = 0.5$  is significantly higher than at the interface.

Near-field polarization of the "extraordinary" SPP in the low-field phase demonstrates very peculiar behaviour. For the spectral range from 506 nm to 711 nm polarization ellipse tilt changes its sign inside the ME crystal. At the wavelength of



Fig. 2. (a) The schematic representation of the corresponding tilt of the polarization ellipse for the low-field (LF) and spin-flop (SF) phases; (b–d) the dependence of the cross-section polarization of the ME SPP ( $\lg |E_y/H_y|$ )) on wavelength in the cases of: (b) the spin-flop phase, extraordinary SPP; (c) the low-field phase, extraordinary SPP; (d) the low-field phase, ordinary SPP.

712 nm the polarization of the SPP is purely TM exactly at the metal-dielectric interface and is twisting with the distance inside  $Cr_2O_3$ . Such polarization behaviour inside  $Cr_2O_3$  described by (15), (16) is a result of the difference in localization coefficients for TM and TE components.

## C. SPP Polarization in the Spin-Flop Phase of $Cr_2O_3$

The polarization ellipse tilt can be observed in the spin-flop  $(\mathbf{L} \perp c$ -axis) phase of  $\operatorname{Cr}_2\operatorname{O}_3$ , for example, in the configuration  $\beta \parallel \mathbf{L}, c$ -axis  $\perp \Xi$ . Comparison of (2) and (3) reveals that in this configuration the emerged toroidal moment  $\tau \parallel \Xi$  and  $\tau \perp \beta$  (see Table I).

The TE/TM ratio in this configuration can be described as follows:

$$\frac{E_y}{H_y} = -i \frac{\lambda_2}{e^{-\gamma_{\rm TM}|z|}} \cdot \left[ \frac{\gamma_{\rm TM}}{\varepsilon_\perp \beta} e^{-\gamma_{\rm TM}|z|} + \frac{1}{\varepsilon_\perp \beta} \frac{\varepsilon_\perp - \gamma_{\rm TM} \gamma_m}{\gamma_m + \gamma_{\rm TE}} e^{-\gamma_{\rm TE}|z|} \right]$$
(17)

The polarization ellipse experiences a slight turn around Oz axis with respect to the non-ME case, see Fig. 2(a) ( $E_y$  and  $E_x$  are in-phase). At the same time, the SPP polarization is inhomogeneous in its cross-section analogous to the case of the low-field phase [see Fig. 2(b)] due to the difference between  $\gamma_{\rm TE}$  and  $\gamma_{\rm TM}$ .

## D. SPP Dispersion and Non-reciprocity in the Spin-flop Phase of $Cr_2O_3$

For definiteness we consider the following configuration: the *c*-axis  $\perp \Xi$ ,  $\beta \uparrow \uparrow Ox$ ,  $\mathbf{L} \uparrow \uparrow Oy$ . At this, the toroidal moment  $\tau \parallel \beta$  and according to Table I in this case one may observe the non-reciprocal propagation constant variation due to the ME



Fig. 3. The ME-induced variation of the SPP dispersion (a) for the spin-flop phase of  $Cr_2O_3$ , and (b) for the material with  $\alpha = 0.08$  (see details in the text). Red and black solid lines correspond to silver and gold interfaces, respectively. Dashed lines correspond to surface plasmon frequencies  $\omega_{sp}$  and dash-dot line shows the asymptote for  $\Delta\beta/\beta$ .

effect. For the SPP propagating along the positive direction of Ox axis the propagation constant has the form

$$\beta = \sqrt{\frac{\varepsilon_m \varepsilon_{\parallel} (\varepsilon_m - \varepsilon_{\perp})}{\varepsilon_m^2 - \varepsilon_{\parallel} \varepsilon_{\perp}}} - \lambda_2 \frac{\varepsilon_m^2}{\varepsilon_m^2 - \varepsilon_{\parallel} \varepsilon_{\perp}}$$
(18)

while opposite  $\beta$  would have the opposite sign of ME contribution to the propagation constant  $\Delta\beta$  (the second term in (18)). This nonreciprocity  $\Delta\beta/\beta$  for the considered configuration is presented in Fig. 3(a). Note that  $\Delta\beta/\beta$  increases at the wavelengths near the surface plasmon frequency  $\omega_{\rm sp}$  and has an asymptote  $\Delta\beta/\beta = \lambda_2/\sqrt{\varepsilon_{\parallel}}$  for higher wavelengths.

 $Cr_2O_3$  possesses rather low values of the ME constants that are of about  $10^{-4}$ . Recently artificial structures have been constructed that exhibit ME properties with the characteristic value of 0.1 so all the effects described in the present paper are significantly enhanced. An example is shown in Fig. 3(b) where the giant non-reciprocity, i.e., ME-induced SPP dispersion variation, is shown for the material with  $\epsilon = 5.5$  and  $\alpha = 0.08$ . These parameters correspond to the BaTiO<sub>3</sub>/BiFeO<sub>3</sub> superlattice fabricated in [28]. The ME nonreciprocity is about  $10^{-2}$ that is three orders higher than for  $Cr_2O_3$  [see Fig. 3(a)].

#### E. Far-field Control Using ME SPPs

The analysis performed above shows how the ME properties of the material provide the variations of the SPP near-field properties. These variations can also be observed in the far-field as the variation of the reflectance spectra of the ME plasmonic structure. Namely, there are two straightforward ways to get the far-field response from the SPP modulation:

1) Dispersion-based measurements: This scheme can be implemented only in the spin-flop phase  $\beta \perp \mathbf{L} \perp c$  (see Table II) so that ME coupling changes the propagation constant of the SPP. The shift of the SPP resonance position is determined by the SPP dispersion variation due to application of the external magnetic field **H** which causes the spin re-orientation in ME crystal. This shift results in the variation of the angular reflectance spectra which can be measured as

$$\delta = \frac{R(\mathbf{H}) - R(0)}{R(\mathbf{H}) + R(0)}.$$
(19)

This idea is very similar to the well-known transverse magnetooptical Kerr effect in magnetic plasmonic structures [51].



Fig. 4. Reflectance angular spectra (blue and red curves for low-field and spin-flop phase, correspondingly) and its variation  $\delta$  (green curve) due to ME switching calculated (a) for  $Cr_2 O_3$ , and (b) for the material with  $\alpha = 0.08$  (see details in the text).

For example, ME SPPs excited at 800 nm wavelength using Kretchmann scheme via GaP prism at the  $Cr_2O_3/gold$  interface in low-field and spin-flop phase provide relative reflectance modulation  $\delta = 13\%$ , see Fig 4(a), and thus can be measured using ordinary experimental techniques. For  $BaTiO_3/BiFeO_3$ superlattice reported in [28] incorporated instead of  $Cr_2O_3$  in the same scheme, relative modulation depth is up to 100%, see Fig. 4(b). Such scheme with ME SPPs can be used not only for efficient modulation of the signal, but for the magnetic-field controlled deflection of the laser beams as well.

2) Polarization-based measurements: Since in certain cases (see Table II) a tilt of SPP polarization ellipse is observed, one can measure this tilt using either polarization or reflectance measurements. Namely, a rotation of the polarization of the reflected light can be measured (which is similar to the magnetooptical Faraday effect) or variation of the reflectance spectra due to the excitation efficiency change of the SPP with tilted polarization ellipse. Moreover, such ME SPPs can be excited using TE-polarized incident light thus providing the possibility for imaging of the spin-flop effect. However the observed polarization-based far-field effects depend on the square of the ME constant and are less pronounced than the dispersion-based measurements.

## IV. CONCLUSION

We have performed the original thorough research of the ME influence on the SPPs. We have shown how one can predict and qualitatively analyze the variations of SPP characteristics in the particular structure only by the form of its ME tensor. We have applied this results to the important special case of the metal/ $Cr_2O_3$  interface.

Two revealed nonreciprocal ME effects are of prime importance for managing of the optical response of the plasmonic structures. The first one is locking between the SPP propagation direction and the polarization ellipse tilt direction due to nonreciprocity, similar to the spin-momentum locking for electronic surface states. The second one is nonreciprocal variation of the SPP propagation constant. It is promising for active control of the far-field optical response of the plasmonic structure by switching of the external magnetic field. Additionally, this phenomenon can be used to observe the phase transitions from the low-field to the spin-flop phase. Using the materias with high ME response in the plasmonic structure, one may perform the magnetic-field controlled deflection of the laser beams as well.

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