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Spin Angular Momentum Transfer and Plasmogalvanic Phenomena

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Abstract. We show that spin angular momentum (SAM) of light is transferred to matter together with momentum and energy, and obtain the expression for the corresponding torque. This can lead to appearance of spin forces, which correspond to the discrepancy between the Lorentz and Eistein-Laub forces. In plasmonic metals these spin forces result in a plasmogalvanic phenomenon, which is pinning of the plasmon-induced electromotive force to atomically-thin layer at the metal interface.

The field of plasmonics studies surface plasmon polaritons (SPPs), which are elementary collective excitations of metal nanostructures which has attracted much attention and found numerous applications over the past several decades. The plasmon drag effect (PLDE), a new non-linear phenomenon, is the giant enhancement of photoinduced rectified electric currents in metal films and nanostructures under surface plasmon resonance conditions [1-13]. This phenomenon is important for applications ranging from plasmonic based electronics to sensing and optoelectronics. PLDE is not fully explained yet and is very interesting from the fundamental point of view as an example of light-matter interaction in strongly enhanced and specially structured plasmonic fields. Here we discuss PLDE starting from general relationships between the conserved quantities in electromagnetic field, such as energy, momentum, angular momentum and rectified responses of materials (electric currents, emf), and apply them, as a particular example, to surface plasmon polaritons (SPPs) in flat geometry.

The conserved quantities have been exhaustively studied in classical mechanics and thermodynamics [14,15]. However the conservation laws for Maxwell equations are well established only in free space for energy $W = \frac{1}{8\pi}(E^2 + H^2)$ [16-18], momentum $\boldsymbol{S} = \frac{1}{8\pi}\boldsymbol{E}\times\boldsymbol{H}$ [16-18], and angular momentum $\boldsymbol{J} = \boldsymbol{r}\times\boldsymbol{S}$ [18]. Recent studies of photonic conserved quantities in free space demonstrate separation of light angular momentum into orbital angular momentum (OAM) and SAM [19,20], conservation laws for angular momentum [21], OAM, and SAM [22]. In interaction with materials, the conserved quantities of electromagnetic fields are not fully understood and are subjects of active debate [23]. Transition from photonic conserved quantities in free space to those in media is crucial and is equivalent to for example introducing potential energy into the full energy of a point particle $E = mv^2/2 + U(x)$ [14]. Here we introduce a new quantity Σ and show that it is a conserved quantity of Maxwell equations in a generic material (including chiral, magnetic, anisotropic media, etc.). We call this conserved quantity "spin" since in free space limit it becomes the electromagnetic spin introduced in 2009 by M. V. Berry [19].

Another aspect not present in free space is transfer of conserved quantities between electromagnetic fields and matter, which for point massive particles is equivalent to presence of Coulomb's surface friction or fluid resistance. Directly from the conservation laws we obtain here, one can see how the SAM of light is transferred to matter, i.e. via the torques provided by Eqs. (3)-(4) below. In classical metal, this torque can be represented in the form of extra "spin" forces applied to the material (see Fig. 1(d)). These forces enter the equation for transfer of the momentum (Eq. (7) below). Note that our result brings more clarity into Lorentz vs. Einstein-Laub forces debate [24-29] and the force distribution paradox presented by

Mansuripur [30], by showing that the discrepancy in the Lorentz and Eistein-Laub force distributions corresponds to transfer of SAM of light.

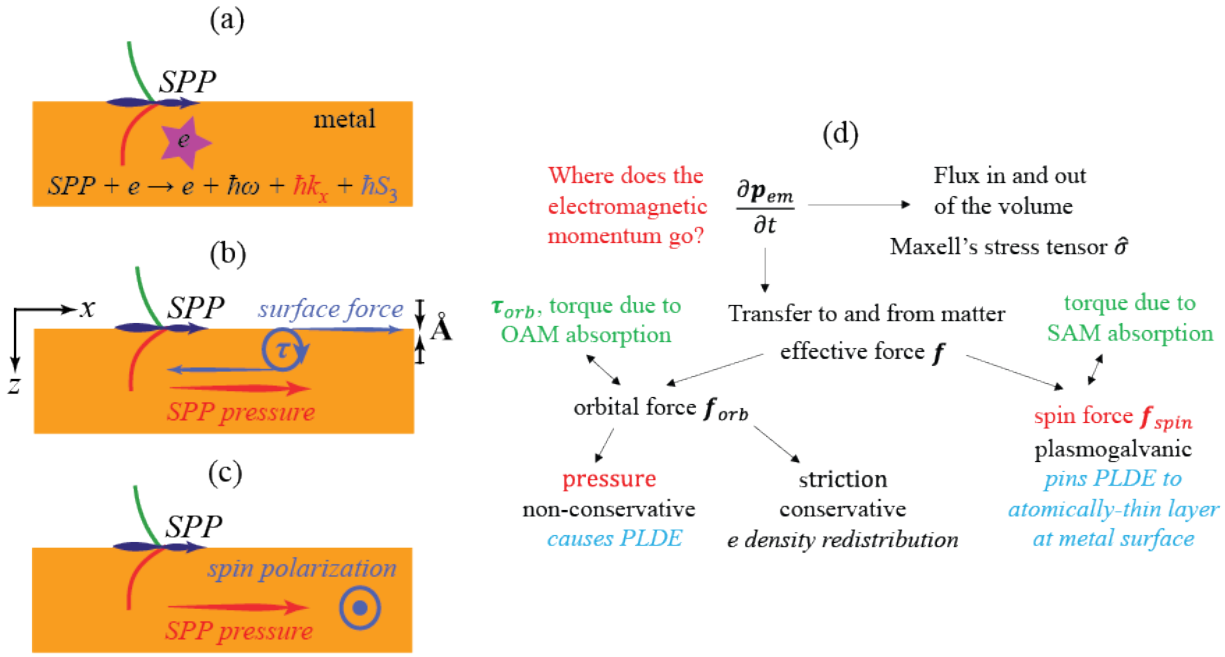

Fig. 1. (a) Schematic of the absorption of an SPP quantum by an electron involving transfer of energy $\hbar\omega$, momentum $\hbar k_x$ and average SAM $\hbar S_3$, where S_3 is the Stokes parameter, characterizing the helicity. (b) Schematic of the plasmon drag pinning to the atomic layer at the metal surface due to SAM absorption. The volume spin forces practically cancel SPP pressure and most of the SPP momentum is deposited by the surface force at the metal interface. (c) Possible scenario when SAM absorption torque results in spin polarization instead of spin forces. (d) Channels for electromagnetic momentum transfer in media and corresponding photoinduced electric effects.

The conserved quantities of electromagnetic fields are bilinear forms of the field vectors. This means that the conserved quantities of continuous wave or narrow-bandwidth pulsed fields are transferred to matter via rectified and second-harmonic channels. The rectified part of the transfer corresponds to the systematic changes in the materials induced by light and PLDE is a major example of such rectification. In metal nanostructures with smooth surfaces the PLDE emf is proportional to the momentum of the surface plasmon polaritons (SPP) and absorbed SPP energy [4]. Irregular or strongly nanostructured surfaces allow for additional contributions into PLDE, which we call *plasmogalvanic effects* [31-37]. Recently the subject of angular momentum of light [38] and its transfer to matter [39,40] have added to the list of topics which can be studied via photo-induced electric responses in metals, in particular by considering currents induced by circularly polarized light off surface plasmon resonance frequncy [12,13].

The SAM of SPPs has attracted considerable attention due to the recent discovery of spin-momentum locking in SPP waves [41-44]. Here, considering generalized fundamental conservation laws for light in matter, we show that SAM of SPPs is absorbed by the metal plasma together with energy and momentum of SPPs (see Fig. 1(a)). From classical point of view, as discussed below, the SAM absorption torque corresponds to additional forces, and results in dramatic redistribution of SPP-induced forces on electrons with localization of those forces at the very surface of metal (Fig. 1(b)). Note that in metals with strong spin-orbital (SO) interaction of electrons such as gold, the SAM absorption may lead to electron spin polarization and corresponding magnetization of the skin-depth layer. In this scenario, the effective force

will be represented by the pressure force distributed over the skin-depth layer, without localization of forces on the metal surface (Fig. 1(c)).

Let us start with the electromagnetic spin conservation law in a lossy material. The conservation law in a lossless dielectric was derived in Ref. [22]. Here, we adopt the following dual-symmetric definition of the SAM of electromagnetic field $\boldsymbol{\Sigma} = \boldsymbol{\Sigma}_e + \boldsymbol{\Sigma}_m$, where $\boldsymbol{\Sigma}_e = \frac{1}{4\pi c}\boldsymbol{E}\times\boldsymbol{A}$ and $\boldsymbol{\Sigma}_m = \frac{1}{4\pi c}\boldsymbol{H}\times\boldsymbol{C}$ [20,22,45], where we define the usual vector potential as $\boldsymbol{B} = \nabla\times\boldsymbol{A}$ and introduce a potential according to $\boldsymbol{D} = -\nabla\times\boldsymbol{C}$ (see also Ref. [46,47]), in both cases using the solenoidal nature of $\boldsymbol{D} = \boldsymbol{E} + 4\pi\boldsymbol{P}$ and $\boldsymbol{B} = \boldsymbol{H} + 4\pi\boldsymbol{M}$. Note that as was shown in Ref. [20] the requirement of the photonic spin to be gauge-invariant corresponds to setting $\nabla\cdot\boldsymbol{A} = 0$ and $\nabla\cdot\boldsymbol{C} = 0$. The potentials are related to the fields as $\boldsymbol{E} = -\frac{1}{c}\frac{\partial\boldsymbol{A}}{\partial t}$ and $\boldsymbol{H} = -\frac{1}{c}\frac{\partial\boldsymbol{C}}{\partial t}$. In monochromatic fields the average over an optical period SAM is $\overline{\boldsymbol{\Sigma}}_e = -\frac{1}{8\pi\omega}\mathrm{Im}\{\boldsymbol{E}\times\boldsymbol{E}^*\}$ and $\overline{\boldsymbol{\Sigma}}_m = -\frac{1}{8\pi\omega}\mathrm{Im}\{\boldsymbol{H}\times\boldsymbol{H}^*\}$.

Consider the time derivative of the spin $\boldsymbol{\Sigma}$

$$\begin{aligned}\frac{\partial\boldsymbol{\Sigma}}{\partial t} &= \frac{1}{4\pi c}\frac{\partial}{\partial t}(\boldsymbol{E}\times\boldsymbol{A} + \boldsymbol{H}\times\boldsymbol{C}) = \frac{1}{4\pi c}\left(\frac{\partial\boldsymbol{E}}{\partial t}\times\boldsymbol{A} + \frac{\partial\boldsymbol{H}}{\partial t}\times\boldsymbol{C}\right) + \frac{1}{4\pi c}\left(\boldsymbol{E}\times\frac{\partial\boldsymbol{A}}{\partial t} + \boldsymbol{H}\times\frac{\partial\boldsymbol{C}}{\partial t}\right)\\ &= \frac{1}{4\pi c}\left(\left(c\nabla\times\boldsymbol{H} - 4\pi\frac{\partial\boldsymbol{P}}{\partial t}\right)\times\boldsymbol{A} + \left(-c\nabla\times\boldsymbol{E} - 4\pi\frac{\partial\boldsymbol{M}}{\partial t}\right)\times\boldsymbol{C}\right)\\ &= \frac{1}{4\pi}(-(\nabla\times\boldsymbol{E})\times\boldsymbol{C} + (\nabla\times\boldsymbol{H})\times\boldsymbol{A}) - \frac{1}{c}\left(\frac{\partial\boldsymbol{P}}{\partial t}\times\boldsymbol{A} + \frac{\partial\boldsymbol{M}}{\partial t}\times\boldsymbol{C}\right) = -\left(\nabla\cdot\hat{\delta}\right) - \boldsymbol{\tau}\end{aligned}$$

This equation has the form of the continuity equation for electromagnetic SAM

$$\frac{\partial\boldsymbol{\Sigma}}{\partial t} + \left(\nabla\cdot\hat{\delta}\right) = -\boldsymbol{\tau}, \tag{1}$$

where $\hat{\delta}$ is the tensor of SAM flux, analogous to Maxwell stress tensor for the momentum flux (cf. Eq. (3.24) of Ref. [22])

$$\hat{\delta} = \left\{\frac{1}{4\pi}\left(\boldsymbol{C}\otimes\boldsymbol{E} - \frac{(\boldsymbol{C}^c\cdot\boldsymbol{E})}{2}\hat{\mathrm{I}}\right) - \frac{1}{4\pi}\left(\boldsymbol{A}\otimes\boldsymbol{H} - \frac{(\boldsymbol{A}^c\cdot\boldsymbol{H})}{2}\hat{\mathrm{I}}\right)\right\}. \tag{2}$$

and $\boldsymbol{\tau} = \boldsymbol{\tau_e} + \boldsymbol{\tau_m}$ is the torque volume density, which is composed from the torques associated with the interaction between polarization and the electric fields $\boldsymbol{\tau_e}$ and magnetization and the magnetic fields $\boldsymbol{\tau_m}$,

$$\boldsymbol{\tau_e} = \frac{1}{c}\frac{\partial}{\partial t}(\boldsymbol{P}\times\boldsymbol{A}) + \boldsymbol{P}\times\boldsymbol{E}, \quad \boldsymbol{\tau_m} = \frac{1}{c}\frac{\partial}{\partial t}(\boldsymbol{M}\times\boldsymbol{C}) + \boldsymbol{M}\times\boldsymbol{H}. \tag{3}$$

In monochromatic fields the first terms in Eqs. (3) result in zero time-average torque, the time-averaged torque density applied to matter is

$$\overline{\boldsymbol{\tau}} = \frac{1}{2}\mathrm{Re}\{\boldsymbol{P}\times\boldsymbol{E}^* + \boldsymbol{M}\times\boldsymbol{H}^*\}. \tag{4}$$

Now let us discuss the electromagnetic torque in dispersive transparent media, which can be considered in a similar manner to the Brillouin internal energy stored in electromagnetic field in media or Abraham force [17]. The second terms in Eqs. (3) have the same form as in the monochromatic fields, and we focus on the first terms, which are zero in monochromatic fields but play a role in a narrow bandwidth pulse. In this case $\frac{\partial\boldsymbol{P}}{\partial t} = -i\omega\chi_e(\omega)\boldsymbol{E} + \frac{d(\omega\chi_e)}{d\omega}\frac{\partial\boldsymbol{E}_0}{\partial t}e^{-i\omega t}$ and $\frac{\partial\boldsymbol{M}}{\partial t} = -i\omega\chi_m(\omega)\boldsymbol{H} + \frac{d(\omega\chi_m)}{d\omega}\frac{\partial\boldsymbol{H}_0}{\partial t}e^{-i\omega t}$, $\boldsymbol{E}_0(t)$ and $\boldsymbol{H}_0(t)$ are slowly varying amplitudes of the fields. In this situation

$$\boldsymbol{\tau} = \frac{1}{c}\frac{\partial}{\partial t}(\boldsymbol{P}\times\boldsymbol{A} + \boldsymbol{M}\times\boldsymbol{C}) == \frac{1}{4c}\left(\frac{\partial \boldsymbol{P}^*}{\partial t}\times\boldsymbol{A} + \frac{\partial \boldsymbol{P}}{\partial t}\times\boldsymbol{A}^* + \frac{\partial \boldsymbol{M}^*}{\partial t}\times\boldsymbol{C} + \frac{\partial \boldsymbol{M}}{\partial t}\times\boldsymbol{C}^*\right) - \frac{1}{2}\mathrm{Re}\{\boldsymbol{P}\times\boldsymbol{E}^* - \boldsymbol{M}\times\boldsymbol{H}^*\} =$$

$$= \frac{1}{4c}\frac{d(\omega\chi_e)}{d\omega}\left(\frac{\partial \boldsymbol{E}_0^*}{\partial t}\times\left(-i\frac{c}{\omega}\boldsymbol{E}_0\right) + \frac{\partial \boldsymbol{E}_0}{\partial t}\times\left(i\frac{c}{\omega}\boldsymbol{E}_0^*\right)\right) + \frac{1}{4c}\frac{d(\omega\chi_m)}{d\omega}\left(\frac{\partial \boldsymbol{H}_0^*}{\partial t}\times\left(-i\frac{c}{\omega}\boldsymbol{H}_0\right) + \frac{\partial \boldsymbol{H}_0}{\partial t}\times\left(i\frac{c}{\omega}\boldsymbol{H}_0^*\right)\right) =$$

$$= -\frac{1}{4\omega}\frac{d(\omega\chi_e)}{d\omega}\frac{\partial}{\partial t}\mathrm{Im}\{\boldsymbol{E}\times\boldsymbol{E}^*\} - \frac{1}{4\omega}\frac{d(\omega\chi_m)}{d\omega}\frac{\partial}{\partial t}\mathrm{Im}\{\boldsymbol{H}\times\boldsymbol{H}^*\} = 2\pi\frac{d(\omega\chi_e)}{d\omega}\frac{\partial\overline{\boldsymbol{\Sigma}_e}}{\partial t} + 2\pi\frac{d(\omega\chi_m)}{d\omega}\frac{\partial\overline{\boldsymbol{\Sigma}_m}}{\partial t}$$

Which means that in narrow-bandwidth pulses in dispersive transparent media with electric and magnetic susceptibilities χ_e and χ_m the Eqs. (3) lead to torque according to

$$\boldsymbol{\tau} = 2\pi\frac{d(\omega\chi_e)}{d\omega}\frac{\partial\overline{\boldsymbol{\Sigma}_e}}{\partial t} + 2\pi\frac{d(\omega\chi_m)}{d\omega}\frac{\partial\overline{\boldsymbol{\Sigma}_m}}{\partial t}.$$

Below we focus on monochromatic fields. In monochromatic fields both expressions for electric and magnetic torques in Eq. 4 can be understood as torques acting on dipole moments in electric and magnetic fields. The expression for the torque $\boldsymbol{\tau_e}$ applied to polarized matter has been used in the proposal of optical torque wrench devices [40], but the role of SAM absorption in matter has never been previously discussed (see also Refs. [48-52]). To show the relation of torque and SAM absorption explicitly, let one assume the material relationship of the form $\boldsymbol{P} = \chi_e\boldsymbol{E} + \xi_e\boldsymbol{H}$ and $\boldsymbol{M} = \chi_m\boldsymbol{H} + \xi_m\boldsymbol{E}$, and the torque in Eq. (4) turns into

$$\overline{\boldsymbol{\tau}} = 4\pi[\omega\chi_e''\overline{\boldsymbol{\Sigma}_e} + \omega\chi_m''\overline{\boldsymbol{\Sigma}_m} + (\xi_m' - \xi_e')\overline{\boldsymbol{S}} + (\xi_e'' + \xi_m'')\tilde{\boldsymbol{S}}]\,, \tag{5}$$

where $\overline{\boldsymbol{S}} = 1/(8\pi)\mathrm{Re}\{\boldsymbol{E}\times\boldsymbol{H}^*\}$ is the Poynting vector averaged over the optical period and $\tilde{\boldsymbol{S}} = -1/(8\pi)\mathrm{Im}\{\boldsymbol{E}\times\boldsymbol{H}^*\}$. Additionally, it appears that loss in bi-anisotropic media, or $\xi_m \neq \xi_e^*$ [53], can result in torque. This fact is of interest due to recent investigations of optical forces applied to bi-anisotropic particles [54-57].

Consider SPP propagation along a flat metal-dielectric interface with $\mu = 1$. The complex fields of a SPP in the metal ($z > 0$) are

$$\boldsymbol{H} = \hat{\boldsymbol{y}}\,e^{-\xi z}e^{i(kx-\omega t)},\quad \boldsymbol{E} = \frac{1}{k_0\varepsilon_m}(-\hat{\boldsymbol{z}}k_x + \hat{\boldsymbol{x}}i\xi)e^{-\xi z}e^{i(kx-\omega t)},\ \frac{\xi}{\varepsilon_m} = -\frac{\kappa_d}{\varepsilon_d}. \tag{6}$$

The last equation is the condition of SPP existence, ensuring the matching of longitudinal electric fields at the metal-dielectric interface, which leads to the dispersion $k_{SPP}(\omega)$ of SPPs (see Fig. 2(a) and Ref [58]).

If we substitute the SPP fields [Eq. (6)] into Eq. (5), which describes SAM absorption, the corresponding torque turns into $\overline{\boldsymbol{\tau}} = -\chi''\mathrm{Im}\{E_xE_z^*\}\hat{\boldsymbol{y}}$. It is important to compare this torque with the energy absorption rate $\bar{Q} = -\frac{\omega}{2}\mathrm{Im}\{P_\alpha E_\alpha^*\}$ as was done in Ref. [4] for momentum. We show in Supplemental Material 1 [60] that the relationship between the torque and energy absorption rate is $\overline{\boldsymbol{\tau}} = \frac{\hbar S_3}{\hbar\omega}\bar{Q}$, where $S_3 = \frac{2\xi k_x}{(k_x^2+\xi^2)}$ is the Stokes parameter, characterizing the helicity (or the degree of circular polarization) in electromagnetic field [42,44,59]. Considering the results of Ref. [4] this means that with absorption of SPP energy quantum $\hbar\omega$ and momentum quantum $\hbar k_x$ electrons gain $\hbar S_3$ amount of angular momentum on average due to absorption of SAM (see Fig. 1(a)).

Electrons have two degrees of freedom, which can accumulate angular momentum: orbital (translational) motion and electron spin. Induction of orbital motion of electrons via absorption of SAM of SPPs corresponds to photoinduced electric currents. At the same time, SAM absorption can lead to change of

electron spin and result in spin polarization as shown in Fig 1(c). Despite large spin-orbit coupling ~ 1 eV [61] and a relatively long spin relaxation time ~ 50 ps [62], spintronic effects in plasmonic materials are not studied. Simultaneous measurement of SPP-generated emf and spin polarization (via for example, pump-probe magneto-optical experiments) in different plasmonic materials could show how angular momentum of light is distributed between the spin of electrons and their translational motion.

Below we consider the implications of the SAM transfer from the electromagnetic field exclusively into the orbital motion of electrons. We start with the momentum transfer, which is described by (see Supplemental Material 2 [60])

$$\frac{\partial \boldsymbol{p}}{\partial t} + \nabla \cdot \hat{\sigma} = -\boldsymbol{f},$$

where $\boldsymbol{p} = 1/(4\pi c)(\boldsymbol{E}\times\boldsymbol{H})$ is the momentum, $\hat{\sigma}$ is the Maxwell stress tensor [16], and $\boldsymbol{f}$ is the volume density of the effective force acting in the medium. The effective force is reminiscent of both Lorentz and Einstein-Laub forces [23-30,50-52,63-66], and is identical to the Lorentz force for $\boldsymbol{M} = 0$, which was previously applied to PLDE (considering only electric responses) in Refs. [2,4]. Considering separately volume and surface contributions, the volume density of the force $\boldsymbol{f}$ is

$$\boldsymbol{f} = -(\nabla \cdot \boldsymbol{P})\boldsymbol{E} + \frac{1}{c}\frac{\partial \boldsymbol{P}}{\partial t}\times\boldsymbol{H} - (\nabla \cdot \boldsymbol{M})\boldsymbol{H} - \frac{1}{c}\frac{\partial \boldsymbol{M}}{\partial t}\times\boldsymbol{E} = \boldsymbol{f}_A + \boldsymbol{f}^{e}_{orb} + \boldsymbol{f}^{m}_{orb} + \boldsymbol{f}^{ve}_{spin} + \boldsymbol{f}^{vm}_{spin} \tag{7}$$

where

(Abraham force) $$\boldsymbol{f}_A = \frac{1}{c}\frac{\partial}{\partial t}(\boldsymbol{P}\times\boldsymbol{H} - \boldsymbol{M}\times\boldsymbol{E} + 4\pi\boldsymbol{P}\times\boldsymbol{M}) \tag{7a}$$

(orbital electric force) $$\boldsymbol{f}^{e}_{orb} = \nabla_{\boldsymbol{E}}(\boldsymbol{P}^{c} \cdot \boldsymbol{E}) \tag{7b}$$

(orbital magnetic force) $$\boldsymbol{f}^{m}_{orb} = \nabla_{\boldsymbol{H}}(\boldsymbol{M}^{c} \cdot \boldsymbol{H}) \tag{7c}$$

(spin electric volume force) $$\boldsymbol{f}^{ve}_{spin} = -\nabla \cdot (\boldsymbol{P} \otimes \boldsymbol{E}) \tag{7d}$$

(spin magnetic volume force) $$\boldsymbol{f}^{vm}_{spin} = -\nabla \cdot (\boldsymbol{M} \otimes \boldsymbol{H}) \tag{7e}$$

The surface density (after integration of the volume density Eq. (7) across the metal interface) is given by

$$\boldsymbol{f}_s = (\boldsymbol{P} \cdot \hat{\boldsymbol{n}})\boldsymbol{E} + (\boldsymbol{M} \cdot \hat{\boldsymbol{n}})\boldsymbol{H} = \boldsymbol{f}^{se}_{spin} + \boldsymbol{f}^{sm}_{spin} \tag{8}$$

(spin electric volume force) $$\boldsymbol{f}^{se}_{spin} = (\boldsymbol{P} \cdot \hat{\boldsymbol{n}})\boldsymbol{E} \tag{8a}$$

(spin magnetic volume force) $$\boldsymbol{f}^{sm}_{spin} = (\boldsymbol{M} \cdot \hat{\boldsymbol{n}})\boldsymbol{H} \tag{8b}$$

Please notice the symmetry of the force Eq. (7)-(8) with respect to the electric and magnetic responses of materials. The Abraham force [Eq. (7a)] is zero in monochromatic fields, similarly to the first terms in Eqs. (4) or to the electromagnetic energy in media [17], and for pulses in $\boldsymbol{M} = 0$ media was considered in [67]. The forces in Eqs. (7b) and (7c) can be viewed as responsible for momentum transfer and change in the OAM [19-22] (i.e. OAM transfer with torque $\boldsymbol{\tau}_{orb} = \boldsymbol{r}\times\boldsymbol{f}_{orb}$) and can be referred to as the orbital force $\boldsymbol{f}_{orb} = \nabla_{\boldsymbol{E}}(\boldsymbol{P}^{c} \cdot \boldsymbol{E}) + \nabla_{\boldsymbol{H}}(\boldsymbol{M}^{c} \cdot \boldsymbol{H})$ (see Fig. 1(d)). The rectified part of the orbital force can be represented as the sum of striction and pressure forces [2]

$$\overline{\boldsymbol{f}^{e}_{orb}} = \frac{1}{2}\mathrm{Re}\{\nabla_{\boldsymbol{E}}(\boldsymbol{P}^{c} \cdot \boldsymbol{E}^{*})\} = \frac{1}{4}\chi'_{e}\nabla(|\boldsymbol{E}|^{2}) - \frac{1}{2}\chi''_{e}\mathrm{Im}\{\nabla_{\boldsymbol{E}}(\boldsymbol{E}^{c} \cdot \boldsymbol{E}^{*})\}, \tag{9a}$$

$$\overline{\boldsymbol{f}^{m}_{orb}} = \frac{1}{2}\mathrm{Re}\{\nabla_{\boldsymbol{H}}(\boldsymbol{M}^{c} \cdot \boldsymbol{H}^{*})\} = \frac{1}{4}\chi'_{m}\nabla(|\boldsymbol{H}|^{2}) - \frac{1}{2}\chi''_{m}\mathrm{Im}\{\nabla_{\boldsymbol{H}}(\boldsymbol{H}^{c} \cdot \boldsymbol{H}^{*})\}, \tag{9b}$$

The pressure force (the terms with χ'' in Eqs. (9)) is proportional to the wave vector of the electromagnetic field and is the source of the PLDE emf. Note that in the orbital force only the pressure force is doing work on electrons while striction is a potential force and is responsible for redistribution of electron density [4]. In Ref. [4] it was demonstrated that the pressure force is directly related to energy absorption, which is manifested in PLDE experiments [1,3].

The remaining contributions in Eqs. (7)-(8), which are in the focus of this manuscript, can be classified as the spin force (see Fig. 1(d)), whose volume density is $\boldsymbol{f}_{spin}^{v} = \boldsymbol{f}_{spin}^{ve} + \boldsymbol{f}_{spin}^{vm}$ and surface density is $\boldsymbol{f}_{spin}^{s} = \boldsymbol{f}_{s} = \boldsymbol{f}_{spin}^{se} + \boldsymbol{f}_{spin}^{sm}$. As was shown in [2], the force densities in (7d) and (8a) satisfy $\oint \boldsymbol{f}_{spin}^{se} \cdot ds - \int \boldsymbol{f}_{spin}^{ve} \cdot dV = 0$. Similarly, $\oint \boldsymbol{f}_{spin}^{sm} \cdot ds - \int \boldsymbol{f}_{spin}^{vm} \cdot dV = 0$. This means that the spin force does not directly contribute to the total force acting on metal electrons and PLDE emf. Instead, these forces are associated with the torque (see Eqs. (4)-(5)), which can be presented as

$$\int (\boldsymbol{P}\times\boldsymbol{E})dV = \oint\big(\boldsymbol{r}\times\boldsymbol{f}_{spin}^{se}\big)ds + \int\big(\boldsymbol{r}\times\boldsymbol{f}_{spin}^{ve}\big)dV, \tag{10a}$$

$$\int (\boldsymbol{M}\times\boldsymbol{H})dV = \oint\big(\boldsymbol{r}\times\boldsymbol{f}_{spin}^{sm}\big)ds + \int\big(\boldsymbol{r}\times\boldsymbol{f}_{spin}^{vm}\big)dV. \tag{10b}$$

Indeed, one can write an identity

$$\begin{aligned}\partial_j\big(\boldsymbol{r}\times\hat{\boldsymbol{x}}_i E_i A_j\big) &= \big(\hat{\boldsymbol{x}}_j\times\hat{\boldsymbol{x}}_i\big)E_i A_j + \boldsymbol{r}\times\hat{\boldsymbol{x}}_i\big(\partial_j E_i\big)A_j + \boldsymbol{r}\times\hat{\boldsymbol{x}}_i E_i\big(\partial_j A_j\big)\\ &= \boldsymbol{A}\times\boldsymbol{E} + \boldsymbol{r}\times\{(\boldsymbol{A}\cdot\nabla)\boldsymbol{E}\} + \boldsymbol{r}\times\{\boldsymbol{E}\cdot(\nabla\cdot\boldsymbol{A})\} = \boldsymbol{A}\times\boldsymbol{E} + \boldsymbol{r}\times\{\nabla\cdot(\boldsymbol{A}\otimes\boldsymbol{E})\}\end{aligned}$$

From this $\boldsymbol{P}\times\boldsymbol{E} = \partial_j\big(\boldsymbol{r}\times\hat{\boldsymbol{x}}_i E_i P_j\big) - \boldsymbol{r}\times\{\nabla\cdot(\boldsymbol{P}\otimes\boldsymbol{E})\}$ and after integration we get (10a).

Note that despite the fact that the SAM absorption torque is only proportional to χ'' (see Eq. (5)), the corresponding rectified spin forces $\boldsymbol{f}_{spin}$ have both χ' and χ'' contributions

$$\overline{\boldsymbol{f}_{spin}^{ve}} = -\tfrac{1}{2}\mathrm{Re}\{\nabla\cdot(\boldsymbol{P}\otimes\boldsymbol{E}^*)\} = \tfrac{1}{4}\chi_e''\mathrm{Im}\{\nabla\times(\boldsymbol{E}\times\boldsymbol{E}^*)\} - \tfrac{1}{2}\chi_e'\mathrm{Re}\{\nabla\cdot(\boldsymbol{E}\otimes\boldsymbol{E}^*)\} \tag{11a}$$

$$\overline{\boldsymbol{f}_{spin}^{vm}} = -\tfrac{1}{2}\mathrm{Re}\{\nabla\cdot(\boldsymbol{M}\otimes\boldsymbol{H}^*)\} = \tfrac{1}{4}\chi_m''\mathrm{Im}\{\nabla\times(\boldsymbol{H}\times\boldsymbol{H}^*)\} - \tfrac{1}{2}\chi_m'\mathrm{Re}\{\nabla\cdot(\boldsymbol{H}\otimes\boldsymbol{H}^*)\} \tag{11b}$$

The first terms of Eq. (11) $\frac{1}{4}\chi_e''\mathrm{Im}\{\nabla\times(\boldsymbol{E}\times\boldsymbol{E}^*)\}$ and $\frac{1}{4}\chi_m''\mathrm{Im}\{\nabla\times(\boldsymbol{H}\times\boldsymbol{H}^*)\}$ can be understood as absorption of the spin part of the Poynting current $P_{3sp} = \mathrm{Im}\{\nabla\times(\boldsymbol{E}\times\boldsymbol{E}^*)\}$ from Ref. [19].

The spin forces represent the SAM transfer from the electromagnetic field into the orbital motion of electrons in the absence of spin polarization. If SAM of SPPs is entirely absorbed to induce spin polarization, the volume density of the effective force acting on electrons is only the orbital force $\overline{\boldsymbol{f}} = \overline{\boldsymbol{f}_{orb}}$. But if the spin polarization is not produced, the effective force acting on electrons and inducing the translational motion is a combination of the orbital and spin forces $\overline{\boldsymbol{f}} = \overline{\boldsymbol{f}_{orb}} + \overline{\boldsymbol{f}_{spin}}$. Note that the difference between the effective force in Eq. (7), which for $\boldsymbol{M} = 0$ is equal to the Lorentz force, and the Einstein-Laub force is $\overline{\boldsymbol{f}_{spin}}$. Therefore, coupling to an internal angular momentum degree of freedom, such as electron spin, determines which force drives the orbital motion of electrons.

In SPP fields [Eq. (6)] the spin electric volume and surface force densities according to Eqs. 7(d) and 8(a)

$$\overline{\boldsymbol{f}_{spin}^{ve}} = -\tfrac{1}{2}\mathrm{Re}\{\nabla\cdot(\boldsymbol{P}\otimes\boldsymbol{E}^*)\} = -\tfrac{1}{2}\mathrm{Re}\{\partial_z(P_z\boldsymbol{E}^*)\} - \tfrac{1}{2}\mathrm{Re}\{\partial_x(P_x\boldsymbol{E}^*)\} \tag{12a}$$

$$\overline{\boldsymbol{f}_{spin}^{se}} = \tfrac{1}{2}\mathrm{Re}\{P_z\boldsymbol{E}^*\}\Big|_{z=0} \tag{12b}$$

The first term in the $\overline{\boldsymbol{f}_{spin}^{ve}}$ integrated over the cross-section of the metal *perpendicular* to SPP propagation gives the exact opposite of the surface force $\overline{\boldsymbol{f}_{spin}^{se}}$. The second term integrated over the cross-section of the metal *in the direction* of SPP propagation is equal to zero assuming fields decay out for $x \to \pm\infty$. If the fields do not fully decay, this part of the spin force is opposite to the surface force created at the ends of the metal in the x-direction. In any case the second term is due to the decay of SPPs in the direction of propagation and does no overall work on electrons in the case of laminar current [4]. Below we consider only the spin force $\overline{\boldsymbol{f}_{spin_z}^{ve}} = -\frac{1}{2}\mathrm{Re}\{\partial_z(P_z\boldsymbol{E}^*)\}$ and disregard $\overline{\boldsymbol{f}_{spin_x}^{ve}} = -\frac{1}{2}\mathrm{Re}\{\partial_x(P_x\boldsymbol{E}^*)\}$.

The surface part of the spin force $\overline{\boldsymbol{f}_{spin}^{se}}$ [Eq. (12b)] is localized at the surface charge layer of metal, i.e. in a 2d layer with thickness on the order of Ångströms. The study of the surface charge layer between metal and dielectric has been a major research direction of metal non-locality in the continuous model [68-70]. Much debate was generated by the proposal that non-local effects at the metal-dielectric interface can be approximated by introduction of an anisotropic transition layer [71-73]. Here we do not pursue the goal of modeling the non-locality as such, but would like to propose a toy-model of a transitional metal surface layer, which gives a visual idea of how the SAM-absorption torque affects the momentum transfer from SPPs to metal plasma.

We first note that at a metal-dielectric interface the tangential component of electric field E_x is continuous, while D_x is not continuous and changes sign through the interface due to negativity of the dielectric permittivity of metal ε_m. This implies that the dielectric permittivity passes through an epsilon-near-zero (ENZ) transition at the metal-dielectric interface in the longitudinal direction. At the same time the normal component of the electric field E_z has a discontinuity at the interface, such that D_z is continuous. We assume that at the boundary the metal fraction $f(z)$ is gradually changing from 0 to 1 within a $d = 2$ Å layer (see Fig. 2(c)), which corresponds to the typical thickness of the surface charge layer [68-73]. This way the surface charge layer becomes a distributed ENZ-ENP metasurface [74], which has dielectric permittivities $\varepsilon_x(z) = \varepsilon_m f(z) + \varepsilon_d\big(1 - f(z)\big)$ and $\varepsilon_z^{-1}(z) = \varepsilon_m^{-1} f(z) + \varepsilon_d^{-1}\big(1 - f(z)\big)$.

For the ENZ-ENP model we use the following metal fraction function $f(z) = \frac{1}{2} + \frac{1}{2\alpha d}(\ln(\cosh \alpha z) - \ln(\cosh \alpha(d - z)))$, which is shown in Fig. 2(c) (we use $d = 2$ Å and $\alpha = 10$ Å^{-1}). The TM polarized fields of SPP wave at the metal-dielectric boundary with such a metasurface can be written as $H_y = H_y(z)e^{i(kx-\omega t)}$, where function $H_y(z)$ satisfies the following equation in the transition layer

$$-\frac{1}{\varepsilon_x}H_y'' - H_y' \cdot \partial_z\left(\frac{1}{\varepsilon_x}\right) = \left(k_0^2 - \frac{k^2}{\varepsilon_z}\right)H_y$$

We solve this equation and match the results at the boundaries of the ENZ-ENP metasurface to get the wave vector of the resulting SPPs (see the dots in Fig. 2(a), which follow very closely the dispersion of SPPs in local model Eq. (13)). We find the electric field according to $E_z = -\frac{kH_y}{k_0\varepsilon_z}$ and $E_x = -\frac{i}{k_0}\frac{H_y'}{\varepsilon_x}$.

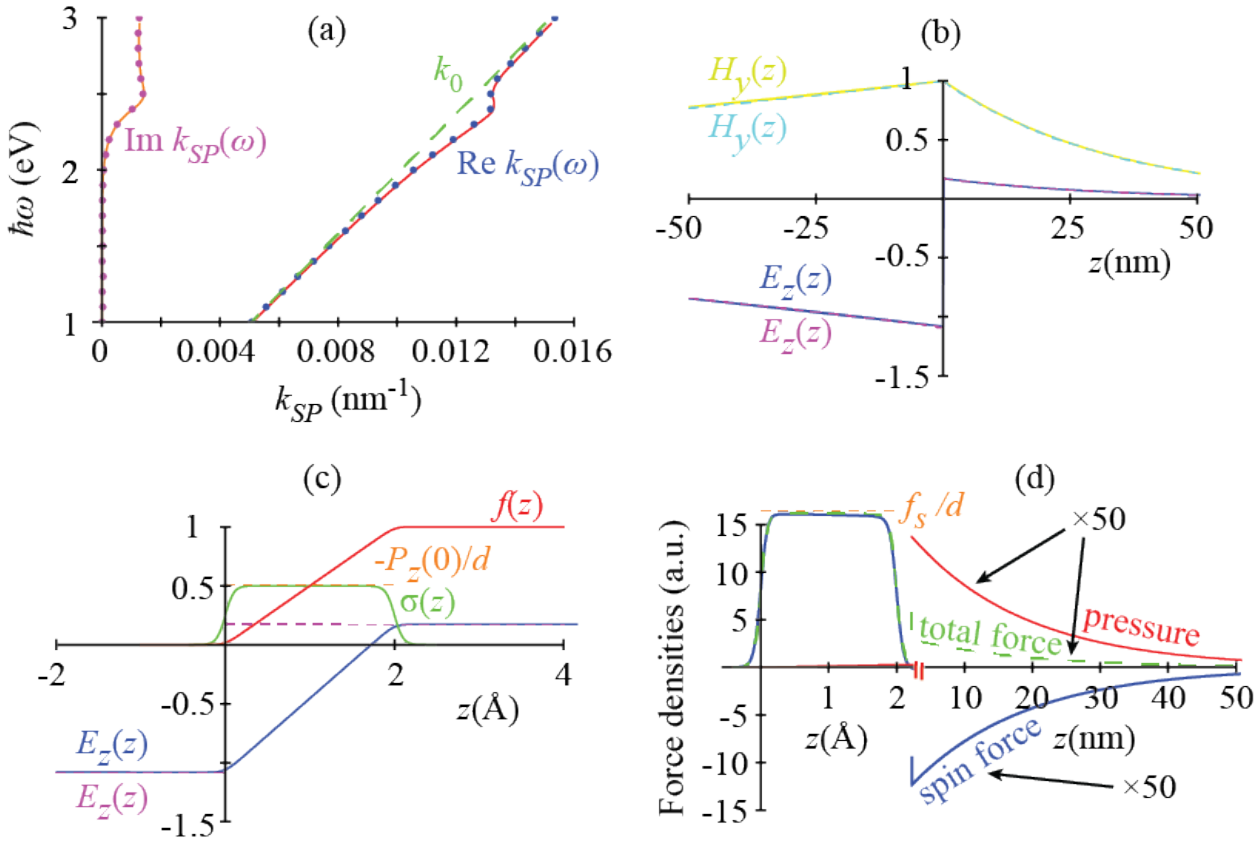

Fig. 2. (a) SPP dispersion in the local model (continuous curves, Eq. (12)) and the ENZ-ENP metasurface model (dots). (b) Comparison of the field distributions in the local model (shown in yellow and blue color) and metasurface approximation (dashed cyan and purple) on the nanoscopic scale. (c) The fields and surface charges in the atomic metasurface region. The metal fraction function $f(z)$ is shown in red. The normal to the interface component of the electric field E_z is shown in blue (metasurface model) and dashed magenta (local model). The surface charge density $\sigma(z)$ is shown in green (metasurface model) and dashed orange (local model). (d) The effect of the spin force (blue) on the total momentum transfer from SPP to electrons (dashed green) as compared to considering only the plasmon pressure force (red). For comparison the surface force in the local model is shown in orange. Note that Fig. 2(d) has an axis break separating the surface shown on the Angstrom scale from -0.5Å to 2.2Å and the skin-depth layer in the metal volume shown on the nanometric scale. All of the forces are multiplied by 50 to the right of the axis break on the nanometric scale.

The resulting wave vector of SPPs (dots in Fig. 2(a)) follows the local model [Eq. (12)]. The magnetic field distribution H_y given by yellow line in Fig. 2(b) follows the local model (cyan dashed line) as well. The electric field distribution is shown in Fig. 2(b)-(c). The normal to the surface component E_z is shown on the nanometric scale in Fig. 2(b) in blue and matches the local model (magenta dashed curve). In Fig. 2(c) one can see the gradual transition of E_z from the metal value to the value in the air within the 2 Å metasurface (blue curve) and compare it to the abrupt jump in the local model. In the metasurface model the oscillating surface charge in the SPP excitation $\sigma(z) \approx -P_z(z=0)/d$ is distributed over the metasurface as seen in Fig. 2(c) in green. Its value corresponds to the local model with $\sigma = -P_z$ (dashed orange).

The results in Fig. 2(d) clarify the possible outcomes of the absorption of the SAM of SPPs on the electronic system of the metal. If the absorption results in spin polarization then the force driving the orbital motion of electrons is the pressure force (red in Fig 2(d)). It is localized on the skin-depth scale. In case the absorption of SAM drives exclusively the orbital motion of electrons, the spin force should be

included (blue in Fig. 2(d)). It has a large contribution within the surface charge layer which is consistent with the value of surface force $\overline{\boldsymbol{f}_s}$ in the local model ($\overline{\boldsymbol{f}_s}/d$ is shown as orange dashed line). In the skin depth layer, outside of the surface charge layer, the spin force becomes considerably lower and is comparable to the pressure force, but is opposite in sign. Integration of the spin force over z results in no overall force. The total force on electrons including the spin force is shown as green dashed line and closely follows the surface component of the spin force in the atomic metasurface region, but is 4 times weaker than the pressure force in the metal volume. This shows that absorption of SAM pins the PLDE force to the surface charge layer, making the force in the skin-depth layer very small.

To conclude, we have introduced and studied a new quantity, photonic spin, which is a conserved quantity of macroscopic Maxwell equations a generic material (including chiral, magnetic, anisotropic media, etc.). We showed that this spin is absorbed by media and derived the expression for the torque corresponding to photonic spin absorption. We demonstrated that this torque corresponds to spin forces applied to material, and that the spin forces correspond to the discrepancy between the Lorentz vs. Einstein-Laub forces. In metals the SAM transfer can lead to modifications in the orbital motion of electrons and/or electron spin polarization. If the orbital channel is prevailing, the action of the spin force in the fields of SPPs at metal-dielectric interface leads to pinning of the PLDE forces to the atomically-thin surface charge layer at the metal interface. The approach developed in our paper for estimation of photonic spin transfer can bring new ideas and directions, such as plasmonic effects in materials with spin orbital interaction, where transfer of SAM from light to carriers can result in electron spin polarization.

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