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Macroscopic drift current in the inverse Faraday effect

Riccardo Hertel[1,*] and Manfred Fähnle[2]

[1]*Institut de Physique et Chimie des Matériaux de Strasbourg, Université de Strasbourg, CNRS UMR 7504, Strasbourg, France*

[2]*Max Planck Institute for Intelligent Systems, Heisenbergstr. 3, 70569 Stuttgart, Germany*

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The inverse Faraday effect (IFE) describes the spontaneous magnetization of a conducting or dielectric medium due to irradiation with a circularly polarized electromagnetic wave. The effect has recently been discussed in the context of laser-induced magnetic switching of solids. We analyze analytically the electron dynamics induced by a circularly polarized laser beam within the framework of plasma theory. A macroscopic drift current is obtained, which circulates around the perimeter of the laser beam. The magnetic moment due to this macroscopic current has an opposite sign and half of the magnitude of the magnetic moment that is generated directly by the IFE. This constitutes an important contribution of angular momentum transferred from the wave to the medium and a classical mechanism for the light-induced generation of magnetic fields.

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Experiments have shown that the magnetic order in solids can be controlled by light [1], an effect that has become known as all-optical switching (AOS). While the details leading to AOS are still not fully understood, the inverse Faraday effect (IFE) (see Ref. [2] and references therein) can be expected to play a major role [3]. The IFE is characterized by the chirality-dependent magnetization generated by a circularly polarized electromagnetic wave. In solid-state magnetism, AOS was believed to be a material-specific effect, occurring only in specific rare-earth transition metal compounds [4]. However, recent experiments have demonstrated AOS in a much broader range of magnetic materials [5]. The phenomenon thus seems to be a general feature of light-matter interaction in the case of circularly polarized electromagnetic waves.

Plasma theory has generally proven to be very efficient to study light-matter interaction in metals. It was recently used to describe the IFE [6,7] in metallic particles and has been successfully applied to various other aspects of the interaction between electromagnetic waves and metals [8]. The assumption of a free electron gas is a simple but powerful model for a metal [9]. In this Rapid Communication we employ single-particle plasma theory to derive a microscopic picture of the IFE. Our results hold, of course, for electron plasmas, but the free electron model may be also used to describe the IFE in metals, especially for high frequencies of the electromagnetic wave [10]. According to the textbook by Landau and Lifshitz [10] this holds typically for ultraviolet light or for x-rays. Insofar the application of our theory to the irradiation of metals with visible light is, of course, some approximation when we consider, e.g., AOS. Nevertheless, we think that our findings will be interesting for people who investigate the effect of circularly polarized light on solids, especially for the community discussing AOS.

As an immediate result one obtains that electrons exhibit a gyrating motion in the electric field of the circularly polarized wave, which leads to a magnetic moment $\mathcal{M}_g$. By tracking the time-averaged motion of free electrons in the field of a collimated circularly polarized electromagnetic wave we find that in addition to these microscopic currents, a macroscopic drift current develops, which circulates at the periphery of the irradiated area. This macroscopic electric current generates a further magnetic moment, $\mathcal{M}_d$. The angular momentum of this additional drift current has not been considered so far and it may provide an ingredient in the open question of the balance of angular momentum in AOS processes.

A circularly polarized electromagnetic wave propagating in the z direction can be written in the usual way, $\boldsymbol{E}(t) = \boldsymbol{E}_0 \exp(-i\omega t)$, where i is the imaginary unit, $\boldsymbol{E}_0$ is the amplitude vector, and $\omega = 2\pi\nu$ is the angular frequency of the wave of frequency ν. Here we have set the phase of the wave to zero without loss of generality. Only the real part of $\boldsymbol{E}(t)$ has a physical meaning. We omit the information that the imaginary component is discarded unless a clarification is necessary. Unlike a linearly polarized wave, $\boldsymbol{E}_0$ is a complex vector in the case of a circularly polarized plane wave:

$$\boldsymbol{E}_0 = \begin{pmatrix} E_\perp \\ \pm i E_\perp \\ 0 \end{pmatrix}, \tag{1}$$

where $(+)$ and $(-)$ stand for left and right circular polarization, respectively, $E_\perp$ is the real-valued amplitude of the electric field and the wave propagates in the z direction. The amplitude $\boldsymbol{E}_0^\pm$ of a circularly polarized wave has some specific properties:

$$i\boldsymbol{E}_0 \times \boldsymbol{E}_0^* = \pm 2E_\perp^2 \hat{\boldsymbol{k}} = \pm|\boldsymbol{E}_0|^2 \cdot \hat{\boldsymbol{k}}, \tag{2}$$

where $\hat{\boldsymbol{k}}$ is a unit vector parallel to the wave's propagation direction, the asterisk $*$ denotes complex conjugation, and $|\boldsymbol{E}_0|^2 = \boldsymbol{E}_0 \cdot \boldsymbol{E}_0^*$. The factor two in Eq. (2) is easily understood by considering that for linearly polarized waves the square of the amplitudes is proportional to the intensity of an electromagnetic wave. Here we have a circularly polarized wave, which can be decomposed into *two* orthogonal, linearly polarized, phase-shifted waves, each with amplitude $E_\perp$.

In the high-frequency limit, the response of the electrons in a dielectric or metallic material to an electromagnetic wave corresponds to that of a free charge [10]. The fundamental equation of motion of a nonrelativistic electron with mass m

*Corresponding author: hertel@ipcms.unistra.fr

and charge e is thus

$$\frac{d\boldsymbol{v}}{dt} = \frac{e}{m}\mathrm{Re}\left\{\boldsymbol{E}_0 \exp(-i\omega t)\right\}. \quad (3)$$

In the electric field of a circularly polarized electromagnetic wave, the motion of the charge results from two superimposed sinusoidal and orthogonal oscillations of equal amplitude, phase-shifted by $\pi/2$. Using the complex amplitude according to Eq. (1) yields

$$v_x = \mp\frac{e}{m\omega}E_y, \quad v_y = \pm\frac{e}{m\omega}E_x, \quad (4)$$

if a dynamic equilibrium is assumed, the Lorentz force of the magnetic field of the wave and the emission of electromagnetic waves by an accelerated electron are neglected, and if, moreover, the initial value conditions are chosen so that any velocity component parallel to the z direction, i.e., parallel to $\boldsymbol{k}$, is discarded. The rotating field of a circularly polarized electromagnetic wave hence results in a circular motion of the electrons around the $\boldsymbol{k}$ axis. In the following we omit the symbol Re when discussing the equations of motion; i.e., we always consider only the real part of the electric field. The velocity components describing this gyration fulfill the equation

$$m\frac{d}{dt}\boldsymbol{v}_\perp = e\boldsymbol{v}_\perp \times \boldsymbol{\Omega}, \quad (5)$$

where $\boldsymbol{v}_\perp(t)$ is the instantaneous vector component of the velocity perpendicular to $\hat{\boldsymbol{k}}$ with $d|\boldsymbol{v}_\perp(t)|/dt = 0$ in the dynamic equilibrium. In Eq. (5) the gyrovector

$$\boldsymbol{\Omega} = -\frac{m\omega}{2eE_\perp^2}(i\boldsymbol{E}_0 \times \boldsymbol{E}_0^*) \quad (6)$$

has been introduced, which is either parallel or antiparallel to $\boldsymbol{k}$, depending on the chirality [cf. Eq. (2)]. Equation (5) has the same form as a Lorentzian force, which evidences a similarity between the IFE and the influence of a static magnetic field. But there are important differences. The rotation frequency of the Larmor precession is proportional to the flux density, while the gyro orbits induced by the IFE have the rotation frequency $\omega = 2\pi\nu$ of the wave, irrespective of the wave's amplitude. Moreover, the Larmor radius is inversely proportional to the strength of the magnetic field, whereas the radius r of the electromagnetic gyro orbits is proportional to the wave's amplitude:

$$r = \frac{e}{m\omega^2}E_\perp. \quad (7)$$

Finally, the velocity $v_\perp$ of the circular motion induced by the circularly magnetized electromagnetic waves is

$$|v_\perp| = \frac{eE_\perp}{m\omega}. \quad (8)$$

These differences between the dynamics of electrons in a magnetic field on one side and the IFE-induced motion on the other make it impossible to assign an unambiguous value to an effective magnetic field that could mimic the IFE. We conclude that, in spite of some qualitative similarities, the IFE cannot be described in a simplified way by means of a magnetic field. This does not mean that there is no magnetic field involved at all in the process. According to Biot-Savart's law, the electrons rotating on gyro orbits generate a magnetic field that, summed over all contributing charges, amounts to a macroscopic field. But this magnetic field is the result of the electromagnetic wave-induced electron dynamics and not a field causing the primary electron motion.

The magnetic moment $\boldsymbol{\mu}$ associated with the circular motion of an electron is the current $[I = e\omega/(2\pi)]$ multiplied with the area that it circumnavigates $A = \pi r^2$. The moment is oriented along the rotation axis, i.e., along $\hat{\boldsymbol{k}}$ in this case. With Eqs. (7), (8), and (2), one obtains

$$\boldsymbol{\mu} = \frac{ev_\perp^2}{2\omega}\hat{\boldsymbol{k}} = \frac{e^3}{4m^2\omega^3}(i\boldsymbol{E}_0 \times \boldsymbol{E}_0^*). \quad (9)$$

With the plasma frequency $\omega_p = (e^2 n_e/\epsilon_0 m)^{(1/2)}$ (ϵ_0 is the vacuum permittivity and n_e the electron density) the above equation can be rewritten

$$\boldsymbol{\mu} = \frac{e\epsilon_0\omega_p^2}{4mn_e\omega^3}(i\boldsymbol{E}_0 \times \boldsymbol{E}_0^*). \quad (10)$$

The total magnetization $\boldsymbol{M}$ is obtained by multiplying Eq. (9) with the density n_e of the conduction electrons. The IFE-induced magnetization $\boldsymbol{M} = n_e\boldsymbol{\mu}$ is thus

$$\boldsymbol{M} = \frac{e\epsilon_0\omega_p^2}{4m\omega^3}(i\boldsymbol{E}_0 \times \boldsymbol{E}_0^*). \quad (11)$$

This result, obtained from single-particle motion (see Ref. [11] and references therein), is identical to that derived from a magnetohydrodynamical approximation [2,12,13]. It appears that the IFE can be described entirely within the theory of classical electrodynamics of continuous media. Our treatment, however, only accounts for the orbital angular momentum of the electronic system acquired by the electromagnetic wave. A quantum-mechanical study will be required to consider possible contributions of light-induced changes of the spin angular momentum, as outlined, e.g., by Popova *et al.* [14]. Consequently, we neglect the influence of related changes via spin-orbit coupling. These changes are considerably smaller than the changes due to the modification of the orbital angular momentum.

So far we have assumed a plane wave, neglecting any variation perpendicular to the propagation direction. However, in the case of circularly polarized waves this common model is problematic. Due to its transverse translational invariance, an infinitely extended circularly polarized plane wave carries no angular momentum [15,16]. Rather than a counterintuitive fact, this result should be interpreted as a sign that the model is too simple in this case to capture essential aspects. It suffices to account for a finite lateral size of the beam to restore the well-known quantum mechanical relation between angular momentum, energy, and frequency in the classical limit [15]. Without further discussing the complicated details of angular momentum balance in the IFE, we refine our analysis based on this knowledge by proceeding with a more realistic model for the electromagnetic wave, i.e., a collimated beam of finite waist w.

To determine the total magnetic moment generated by the IFE we perform a volume integral of the magnetization $\boldsymbol{M}(\boldsymbol{r})$ over the region irradiated by the laser spot. The profile of a

Gaussian beam is

$$\boldsymbol{E}_0(r) = \begin{pmatrix} E_\perp(r) \\ \pm i E_\perp(r) \\ 0 \end{pmatrix} = \begin{pmatrix} 1 \\ \pm i \\ 0 \end{pmatrix} E_\perp^0 \exp\left(\frac{-r^2}{w^2}\right), \tag{12}$$

where r is the distance from the beam axis, w is the waist of the beam, and $E_\perp^0 = E_\perp(r=0)$. If this position-dependent amplitude $\boldsymbol{E}_0(r)$ is plugged into Eq. (11), a volume integral yields the total magnetic moment (z is the thickness; we assume homogeneity along z, thereby restricting the analysis to thin films):

$$\begin{aligned}\mathcal{M}_g &= \frac{e\epsilon_0\omega_p^2}{4m\omega^3}\left[i\boldsymbol{E}_0(r=0)\times\boldsymbol{E}_0^*(r=0)\right] \\ &\quad\times\int_{z'=0}^{z}\int_{r=0}^{\infty}\int_{\phi=0}^{2\pi} r\exp\left(-\frac{2r^2}{w^2}\right)drd\phi dz' \\ &= \frac{w^2\pi z e\epsilon_0\omega_p^2}{8m\omega^3}\left[i\boldsymbol{E}_0(r=0)\times\boldsymbol{E}_0^*(r=0)\right]\end{aligned} \tag{13}$$

Note that the result is proportional to $w^2\pi$, i.e., to the size of the laser spot. Microscopically, the collimation of the beam results in a local amplitude gradient perpendicular to the wave's propagation direction. It can be assumed that electrons' IFE-induced gyration radius is much smaller than the characteristic length on which the amplitude changes:

$$\chi = \frac{e|\nabla E_\perp|}{m\omega^2} \ll 1. \tag{14}$$

On the length scale of a gyro radius, the isolines of the field amplitude are almost parallel. In the following we will show that the central point of the gyro orbit, the guiding center [17], moves ("drifts") along these isolines on a time scale much slower than ω^{-1}.

Because the problem is radially symmetric we consider for simplicity that the amplitude gradient is in the y direction. Then the changes of the amplitude $E_\perp$, linearized along the gyration orbit with guiding center at y_{gc}, yield a position-dependent field according to

$$\boldsymbol{E}(y) = \boldsymbol{E}_{\mathrm{gc}} + (y - y_{\mathrm{gc}})\frac{dE_\perp}{dy}\frac{\boldsymbol{E}_{\mathrm{gc}}}{E_\perp}, \tag{15}$$

where $\boldsymbol{E}_{\mathrm{gc}}$ is the electric field at the guiding center. Note that the electromagnetic wave is circularly polarized, everywhere and at any time. But as electrons move along gyro orbits, they periodically enter regions of larger and smaller amplitude. These variations affect the overall motion, and, as we will show, this results in a macroscopic drift current due to the motion of the guiding centers, a current which circulates around the finite perimeter of the laser beam. Small velocity changes are accounted for by expanding $\boldsymbol{v}$ in a perturbation series, where higher-order terms are corrections to the unperturbed motion $\boldsymbol{v}_0$. In the expansion $\boldsymbol{v} = \boldsymbol{v}_0 + \boldsymbol{v}_1 + \boldsymbol{v}_2 + \cdots$ the smallness parameter is given by Eq. (14), meaning that the order of magnitude of the addends in the perturbation series decreases by a factor of χ with each index number. The goal is to separate the orders of magnitudes of $\boldsymbol{v}$, which is effectively a separation of time scales. This will allow us to deduce that a perturbation of the high-frequency motion can induce a slow drift of the guiding center. The starting point is the equation of motion,

$$\frac{d}{dt}mv_x = \pm m\omega v_y \pm m\omega(y - y_{\mathrm{gc}})\frac{1}{E_\perp}\frac{dE_\perp}{dy}v_y, \tag{16}$$

$$\frac{d}{dt}mv_y = \mp m\omega v_x \mp m\omega(y - y_{\mathrm{gc}})\frac{1}{E_\perp}\frac{dE_\perp}{dy}v_x, \tag{17}$$

where the electric field components E_x, E_y have been written in terms of $\boldsymbol{v}$ using Eq. (4). To study the drift motion, the perturbation expansion of $\boldsymbol{v}$ is inserted and a time average $\langle,\rangle$ over several oscillation periods is performed. The zero-order terms are harmonic oscillations and their time-averaged value is zero. The remaining first-order terms are

$$\left\langle\frac{dv_{1,x}}{dt}\right\rangle = \pm\omega\langle v_{1,y}\rangle \pm \frac{\omega}{E_\perp}\frac{dE_\perp}{dy}\langle(y_0 - y_{\mathrm{gc}})v_{0,y}\rangle, \tag{18}$$

$$\left\langle\frac{dv_{1,y}}{dt}\right\rangle = \mp\omega\langle v_{1,x}\rangle \mp \frac{\omega}{E_\perp}\frac{dE_\perp}{dy}\langle(y_0 - y_{\mathrm{gc}})v_{0,x}\rangle. \tag{19}$$

The terms on the left-hand side are proportional to the *change in time* of the first-order correction $\boldsymbol{v}_1$. The time derivatives of first-order terms are *de facto* second-order terms, since they describe changes in time of a motion that is already much slower than $\boldsymbol{v}_0$. Hence, the time derivatives $d\boldsymbol{v}_1/dt$ are negligibly small compared to the individual terms on the right-hand side, leading to

$$\langle v_{1,y}\rangle = -\frac{1}{E_\perp}\frac{dE_\perp}{dy}\langle(y_0 - y_{\mathrm{gc}})v_{0,y}\rangle, \tag{20}$$

$$\langle v_{1,x}\rangle = -\frac{1}{E_\perp}\frac{dE_\perp}{dy}\langle(y_0 - y_{\mathrm{gc}})v_{0,x}\rangle. \tag{21}$$

The term $\langle(y_0 - y_{\mathrm{gc}})v_{0,y}\rangle$ is zero because in the unperturbed gyromotion the oscillations of position and velocity along the y axis are phase-shifted by $\pi/2$, yielding $\langle v_{1,y}\rangle = 0$. A constant drift is obtained from Eq. (21),

$$\langle v_{1,x}\rangle = -\frac{1}{E_\perp}\frac{dE_\perp}{dy}\langle(y_0 - y_{\mathrm{gc}})v_{0,x}\rangle = \pm\frac{v_\perp^2}{2\omega E_\perp}\frac{dE_\perp}{dy}, \tag{22}$$

where $v_\perp = \sqrt{v_x^2 + v_y^2}$.[1]

Using $v_\perp = eE_\perp/(m\omega)$ yields

$$\langle v_{1,x}\rangle = \pm\frac{e^2E_\perp}{2m^2\omega^3}\left(\frac{dE_\perp}{dy}\right). \tag{23}$$

After transition from the local frame, where the amplitude gradient was assumed to be along y, to the general case where the amplitude decays with increasing r, the drift current flows along the isolines of the field amplitude. In a radially symmetric situation, the drift currents are thus azimuthal, along the unit vector $\boldsymbol{e}_\phi$, which is perpendicular to both the amplitude gradient and the propagation direction. The corresponding

[1] Note that $\langle y_{\mathrm{gc}}v_{0,x}\rangle$ can be neglected as y_{gc} is almost constant on the time scale $\tau \simeq \omega^{-1}$ of the time average $\langle,\rangle$, so that $\langle(y_0 - y_{\mathrm{gc}})v_{0,x}\rangle = \langle y_0(t)v_{0,x}(t)\rangle = \mp\langle\frac{1}{\omega}v_{0,x}(t)v_{0,x}(t)\rangle = \mp v_\perp^2/(2\omega)$.

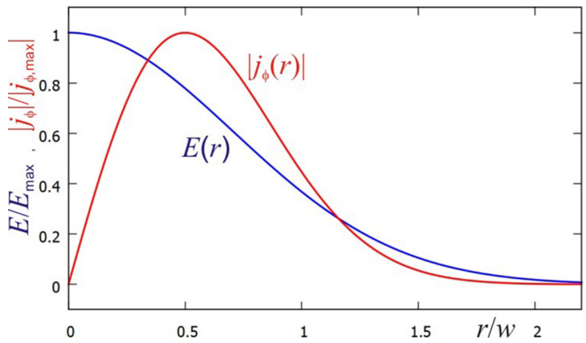

FIG. 1. (Color online) Normalized Gaussian field amplitude $E(r)$ and normalized magnitude of the drift current density $j_\phi(r)$ as a function of the distance r from the center of the beam, measured in units of the beam waist w. The current flows circularly around the $r = 0$ axis with helicity-dependent sign.

current density is

$$\boldsymbol{j}_\phi = ne\langle v_\phi\rangle \boldsymbol{e}_\phi = \pm\frac{n_e e^3 E_\perp}{2m^2\omega^3}\left(\frac{dE_\perp}{dr}\right)\boldsymbol{e}_\phi. \tag{24}$$

The profile Eq. (12) yields a position-dependent current density as shown in Fig. 1:

$$\boldsymbol{j}_\phi(r) = \mp\frac{n_e e^3}{m^2\omega^3 w^2}\left(E_\perp^0\right)^2 r\exp\left(-\frac{2r^2}{w^2}\right)\boldsymbol{e}_\phi \tag{25}$$

Assuming homogeneity along the thickness z, an infinitesimal current flows at radius r according to

$$d\boldsymbol{I}_\phi(r) = \mp\frac{n_e e^3\left(E_\perp^0\right)^2}{m^2\omega^3 w^2} r\exp\left(-\frac{2r^2}{w^2}\right) z dr\cdot\boldsymbol{e}_\phi \tag{26}$$

This current generates an infinitesimal magnetic moment

$$d\boldsymbol{\mathcal{M}}_d = \mp\hat{\boldsymbol{k}}\cdot r^2\pi\cdot\frac{zn_e e^3\left(E_\perp^0\right)^2}{m^2\omega^3 w^2} r\exp\left(-\frac{2r^2}{w^2}\right)dr \tag{27}$$

The index d recalls that this term is due to drift currents. The total magnetic moment is

$$\begin{aligned}\boldsymbol{\mathcal{M}}_d &= \mp\hat{\boldsymbol{k}}\frac{\pi z n_e e^3\left(E_\perp^0\right)^2}{m^2\omega^3 w^2}\int_{r=0}^{\infty} r^3\exp\left(-\frac{2r^2}{w^2}\right)dr\\ &= \mp\frac{w^2\pi z n_e e^3}{8m^2\omega^3}\hat{\boldsymbol{k}}\left(E_\perp^0\right)^2\\ &= -\frac{w^2\pi z e\epsilon_0\omega_p^2}{16m\omega^3}\left[i\boldsymbol{E}_0(r=0)\times\boldsymbol{E}_0^*(r=0)\right]\end{aligned} \tag{28}$$

where Eq. (2) has been used. A comparison with the magnetic moment of the microscopic currents $\boldsymbol{\mathcal{M}}_g$, according to Eq. (13), yields $\boldsymbol{\mathcal{M}}_d = -\boldsymbol{\mathcal{M}}_g/2$.

In conclusion, the circularly polarized laser beam has a twofold effect in the IFE, as it generates two magnetic moment components. These components are collinear, have opposite sign, and differ by a factor of two. Since the system's angular momentum is proportional to its magnetic moment, neglecting the drift current overestimates the IFE-induced angular momentum by a factor of two. Hence, the angular momentum transfer in AOS is much lower than one would expect from a model that assumes translational invariance. This finding could stimulate future studies, especially concerning the open question of angular momentum transfer in the AOS. While our study does not resolve immediately a controversy in the literature, it provides valuable information on the importance of the finite lateral size of a laser beam in the IFE. We have also shown that, contrary to common assumptions, the IFE cannot be described by an effective field.

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