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A relativistic interaction Hamiltonian coupling the angular momentum of light and the electron spin

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On the basis of the Dirac equation, a relativistic interaction Hamiltonian is derived which linearly couples the angular momentum density $\boldsymbol{j}$ of the electromagnetic (EM) field and the electron's spin $\boldsymbol{\sigma}$. The expectation value of this novel Hamiltonian is demonstrated to be precisely the recently proposed energy coupling the EM angular momentum density and magnetic moments [A. Raeliarijaona *et al.*, Phys. Rev. Lett. **110**, 137205 (2013)]. This previously overlooked Hamiltonian is also found to naturally result in the exact analytical form of the interaction energy inherent to the inverse Faraday effect, therefore demonstrating its relevance and easy use for the derivation of other complex magneto-optical and magneto-electric effects originating from electron spin-light angular momentum-couplings.

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A coupling energy between angular momentum density of an electromagnetic (EM) field and magnetic moments in a material has been recently proposed[1], with the following form:

$$\mathcal{E} = \xi \int_{\Omega} [\boldsymbol{r} \times (\boldsymbol{E} \times \boldsymbol{B})] \cdot \boldsymbol{M} \, d\boldsymbol{r}. \qquad (1)$$

where ξ is a material-dependent coefficient, $\boldsymbol{r}$ is the vector position, $\boldsymbol{E}$ is the electric field, $\boldsymbol{B}$ is the magnetic field and $\boldsymbol{M}$ is the magnetization. Here $\boldsymbol{j} = \boldsymbol{r} \times (\boldsymbol{E} \times \boldsymbol{B})$ is the density of total angular momentum of the EM field[2,3]. Surprisingly, such coupling energy has been shown to naturally explain various experimentally known, complex spin-orbit-driven effects. Examples include the spin-current model in multiferroics[1], the anomalous Hall effect[4], planar Hall effect and anisotropic magnetoresistance[5] in ferromagnets. It even led to the prediction of striking spintronics phenomena at interfaces, that bear resemblance to the so-called inverse Rashba-Edelstein-like effects[6].

However, the energy of Eq. (1) was proposed solely based on symmetry arguments and on the control of magnetic vortices by the cross-product between the electric field and magnetic fields[1]. In other words, its physical origin still remains to be elucidated, which will then definitely make it a novel, robust and straightforward predictive tool. One may, for instance, wonder if it is possible to trace Eq. (1) to the most fundamental quantum-mechanical Hamiltonian involving relativistic effects such as the spin-orbit interaction, *viz.* the Dirac equation. If that is the case, it will not only establish the existence of Eq. (1) at a fundamental level, but also lead to the possibility of deriving and understanding *other* spin–angular momentum effects.

In this Communication, we demonstrate that the Dirac Hamiltonian naturally leads to a previously overlooked electron spin–light angular momentum-interaction Hamiltonian. In what follows, we will refer to this term as the Angular MagnetoElectric (AME) coupling Hamiltonian. Quite remarkably, the expectation value of the AME Hamiltonian is found to precisely be Eq. (1) for some materials (e.g., in ferromagnets). Moreover, the use of this general AME Hamiltonian is also shown here to reproduce in a straightforward manner the analytical form of the potential inherent to the so-called and complex inverse Faraday effect[7–9] – which further demonstrates the enormous and broad importance of the AME Hamiltonian.

Derivation of the AME Hamiltonian:
Let us start from the Dirac Hamiltonian for an electron (mass m, charge e) in an electromagnetic field:

$$\mathcal{H}_{\mathrm{D}} = c\, \boldsymbol{\alpha} \cdot (\boldsymbol{p} - e\boldsymbol{A}) + (\beta - \mathbb{1})mc^2 + V + e\Phi, \qquad (2)$$

where $\boldsymbol{A}(\boldsymbol{r}, t)$ is the time-varying EM vector potential, $\boldsymbol{p}$ is the momentum operator, V is the crystal potential, $\Phi(\boldsymbol{r}, t)$ is the scalar potential of the EM field, $\mathbb{1}$ is the 4×4 unit matrix and $\boldsymbol{\alpha}$ and β are 4×4 matrices given, for instance, in Ref.[10]. Applying the Foldy-Wouthuysen transformation[11] to Eq. (2), expanding the result in powers of $1/c$ and keeping terms up to second order, we arrive at the Hamiltonian for the large component of Dirac bi-spinor (see Ref. 12):

$$\mathcal{H}_{\rm FW} = \frac{(\boldsymbol{p}-e\boldsymbol{A})^2}{2m} + V + e\Phi - \frac{e\hbar}{2m}\boldsymbol{\sigma}\cdot\boldsymbol{B} - \frac{(\boldsymbol{p}-e\boldsymbol{A})^4}{8m^3c^2} - \frac{1}{8m^2c^2}\left(p^2V\right) - \frac{e\hbar^2}{8m^2c^2}\nabla\cdot\boldsymbol{E}_{\rm ext}$$
$$+ \frac{i}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{p}V)\times(\boldsymbol{p}-e\boldsymbol{A}) - \frac{e\hbar}{8m^2c^2}\boldsymbol{\sigma}\cdot\{\boldsymbol{E}_{\rm ext}\times(\boldsymbol{p}-e\boldsymbol{A}) - (\boldsymbol{p}-e\boldsymbol{A})\times\boldsymbol{E}_{\rm ext}\}\,. \tag{3}$$

Here $\boldsymbol{B} = \boldsymbol{\nabla}\times\boldsymbol{A}$ is the external magnetic field and $\boldsymbol{E}_{\rm ext} = -\partial\boldsymbol{A}/\partial t - \boldsymbol{\nabla}\Phi$ is the external electric field. The last two terms of Eq. (3) contain the spin-orbit interaction,

$$\mathcal{H}_{\rm SOC} = \frac{i}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{p}V)\times(\boldsymbol{p}-e\boldsymbol{A}) - \frac{e\hbar}{8m^2c^2}\boldsymbol{\sigma}\cdot\{\boldsymbol{E}_{\rm ext}\times(\boldsymbol{p}-e\boldsymbol{A}) - (\boldsymbol{p}-e\boldsymbol{A})\times\boldsymbol{E}_{\rm ext}\}\,. \tag{4}$$

We can re-arrange $\mathcal{H}_{\rm SOC}$ as:

$$\mathcal{H}_{\rm SOC} = \mathcal{H}_{\rm TSOC} + \mathcal{H}_{\rm AME} \tag{5}$$

with

$$\mathcal{H}_{\rm TSOC} = \frac{i}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{p}V\times\boldsymbol{p}) - \frac{e\hbar}{8m^2c^2}\boldsymbol{\sigma}\cdot\{\boldsymbol{E}_{\rm ext}\times\boldsymbol{p} - \boldsymbol{p}\times\boldsymbol{E}_{\rm ext}\}$$
$$= \frac{\hbar}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{\nabla}V\times\boldsymbol{p}) - \frac{e\hbar}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{E}_{\rm ext}\times\boldsymbol{p}) + \frac{ie\hbar^2}{8m^2c^2}\boldsymbol{\sigma}\cdot\partial\boldsymbol{B}/\partial t, \tag{6}$$

$$\mathcal{H}_{\rm AME} = -i\frac{e}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{p}V\times\boldsymbol{A}) + \frac{e^2\hbar}{8m^2c^2}\boldsymbol{\sigma}\cdot\{\boldsymbol{E}_{\rm ext}\times\boldsymbol{A} - \boldsymbol{A}\times\boldsymbol{E}_{\rm ext}\}\,, \tag{7}$$

where $\mathcal{H}_{\rm TSOC}$ contains the "traditional" spin-orbit interaction, and $\mathcal{H}_{\rm AME}$ contains, what we call, the AME interaction. An important point to note is that neither the AME Hamiltonian $\mathcal{H}_{\rm AME}$ nor $\mathcal{H}_{\rm TSOC}$ are gauge invariant. It is rather the total spin-orbit Hamiltonian $\mathcal{H}_{\rm SOC}$ which is gauge invariant. Note that, in the Coulomb gauge, both the magnetic $\boldsymbol{B}$ and magnetic induced electric $\boldsymbol{E}_{\rm ext}$ fields are transverse. Introducing the total electric field $\boldsymbol{E} = \boldsymbol{E}_{\rm ext} - \frac{1}{e}\boldsymbol{\nabla}V$, we can write the AME coupling in a very concise form as

$$\mathcal{H}_{\rm AME} = \frac{e^2\hbar}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{E}\times\boldsymbol{A})\,. \tag{8}$$

Interestingly, the relativistic interaction Hamiltonian adopts the simple form $\zeta\,\boldsymbol{\sigma}\cdot\boldsymbol{j}_s$, where the EM spin angular momentum density[3] is expressed as $\boldsymbol{j}_s = -2\boldsymbol{E}\times\boldsymbol{A}$ ($\epsilon_0 = 1$). This term can be considered as the EM-field analog of the conventional spin-orbit coupling term in $\mathcal{H}_{\rm TSOC}$, which (for central symmetric potentials) gives the well-known interaction $\lambda\,\boldsymbol{\sigma}\cdot\boldsymbol{l}$ with the orbital moment $\boldsymbol{l}$. The existence of this coupling term highlights the appealing possibility to manipulate the spin of electrons in materials by suitably shaped EM waves possessing angular momentum.

The total electric field in a material can be split in terms of an internal electric field $\boldsymbol{E}_{\rm int}$, generated by all charges *in* the material, and an applied electric field $\boldsymbol{E}_{\rm ext}$ which would exist even in vacuum. Of course, the material shall screen the applied electric field. Therefore, the internal electric field depends on $\boldsymbol{E}_{\rm ext}$, which in the limit of small applied field can be dealt with in the framework of linear response theory:

$$\boldsymbol{E}_{\rm int} = \boldsymbol{E}^0_{\rm int} + \overleftrightarrow{\gamma}\cdot\boldsymbol{E}_{\rm ext}, \tag{9}$$

where $\boldsymbol{E}^0_{\rm int} = -\frac{1}{e}\boldsymbol{\nabla}V$ is the internal electric field when no external field is applied and, in general, $\overleftrightarrow{\gamma}$ is a 3×3 matrix. For the sake of simplicity we will assume a cubic or isotropic material, so that $\overleftrightarrow{\gamma}$ reduces to a scalar γ. Therefore, we can re-write the AME coupling according Eqs. (8) and (9) as the sum of an intrinsic part $\mathcal{H}^{\rm int}$ and an induced part $\mathcal{H}^{\rm ind}$:

$$\mathcal{H}_{\rm AME} = \underbrace{\frac{e^2\hbar}{4m^2c^2}\boldsymbol{\sigma}\cdot\left(\boldsymbol{E}^0_{\rm int}\times\boldsymbol{A}\right)}_{\mathcal{H}^{\rm int}_{\rm AME}} + \underbrace{(1+\gamma)\frac{e^2\hbar}{4m^2c^2}\boldsymbol{\sigma}\cdot(\boldsymbol{E}_{\rm ext}\times\boldsymbol{A})}_{\mathcal{H}^{\rm ind}_{\rm AME}}\,. \tag{10}$$

The first term of this Hamiltonian acts as an anisotropy term and will not be given further consideration in what follows. Instead, we will focus on the second term, which is material dependent through the constant γ that characterizes the linear response of the material to an external field. The Hamiltonian of interest is therefore

$$\mathcal{H}^{\rm ind}_{\rm AME} = a\,\boldsymbol{\sigma}\cdot(\boldsymbol{E}_{\rm ext}\times\boldsymbol{A})\;, \tag{11}$$

with $a = (1+\gamma)\frac{e^2\hbar}{4m^2c^2}$.

Relation between the AME Hamiltonian and the previously proposed Eq. (1):

Let us now analyze the case of a homogenous magnetic field inside a magnetic media, which is, e.g., the case for the anomalous Hall effect[4], planar Hall effect and anisotropic magnetoresistance[5] in ferromagnets. Using the Coulomb gauge for a uniform and *slowly varying* magnetic field, the vector potential is given by

$$\boldsymbol{A} = \frac{\boldsymbol{B} \times \boldsymbol{r}}{2}, \tag{12}$$

where $\boldsymbol{r}$ is the position vector. Note that the vector potential is in itself subject to gauge transformations, however, when choosing $\boldsymbol{\nabla} \cdot \boldsymbol{A} = 0$ only the transverse part of $\boldsymbol{A}$ is retained, which is gauge invariant (see, e.g., Ref. 13). The energy related to the induced term of the AME coupling in Eq. (10) can be obtained by taking its expectation value

$$\mathcal{E}^{\rm ind}_{\rm AME} = \xi \int_{\Omega} \boldsymbol{M}(r) \cdot [\boldsymbol{E}_{\rm ext} \times (\boldsymbol{B} \times \boldsymbol{r})] d\boldsymbol{r} \tag{13}$$

where ξ is a material-dependent coefficient, Ω is the volume of the system and $\boldsymbol{M}$ is the magnetization.

With the help of the vector identity $\boldsymbol{a} \times (\boldsymbol{b} \times \boldsymbol{c}) = (\boldsymbol{a} \cdot \boldsymbol{c})\, \boldsymbol{b} - (\boldsymbol{a} \cdot \boldsymbol{b})\, \boldsymbol{c}$, the energy in Eq. (13) can be written as

$$\mathcal{E}^{\rm ind}_{\rm AME} = \xi \int_{\Omega} [(\boldsymbol{M} \cdot \boldsymbol{B})(\boldsymbol{r} \cdot \boldsymbol{E}_{\rm ext}) - (\boldsymbol{M} \cdot \boldsymbol{r})(\boldsymbol{E}_{\rm ext} \cdot \boldsymbol{B})]\, d\boldsymbol{r}. \tag{14}$$

Finally, by employing that $\boldsymbol{M} = \chi_{\rm m} \boldsymbol{H}$, with $\chi_{\rm m}$ the volume magnetic susceptibility, and the linear relation $\boldsymbol{B} = (1 + \chi_{\rm m}^{-1}) \boldsymbol{M}$ ($\mu_0 = 1$), we can interchange $\boldsymbol{B}$ and $\boldsymbol{M}$ in the second term of Eq. (14). This leads to the following expression for the AME energy

$$\begin{aligned}\mathcal{E}^{\rm ind}_{\rm AME} &= \xi \int_{\Omega} [(\boldsymbol{M} \cdot \boldsymbol{B})(\boldsymbol{r} \cdot \boldsymbol{E}_{\rm ext}) - (\boldsymbol{B} \cdot \boldsymbol{r})(\boldsymbol{E}_{\rm ext} \cdot \boldsymbol{M})]\, d\boldsymbol{r} \\ &= \xi \int_{\Omega} [\boldsymbol{r} \times (\boldsymbol{E}_{\rm ext} \times \boldsymbol{B})] \cdot \boldsymbol{M}\, d\boldsymbol{r}. \end{aligned} \tag{15}$$

Remarkably, Equation (15) is precisely the coupling energy between the angular momentum density of the EM field and magnetic moments given in Eq. (1)! One can therefore conclude that the AME Hamiltonian of Eq. (11) is the source of this coupling energy and thus of the different spin-driven magnetoelectric effects that have been recently re-derived via the use of Eq. (1) – such as the spin-current model in multiferroics[1], anomalous Hall effect[4], planar Hall effect and anisotropic magnetoresistance[5] in ferromagnets.

Application of the AME Hamiltonian to the inverse Faraday effect:

Let us now take advantage of the present discovery of this AME Hamiltonian to determine if other magnetoelectic effects can be "easily" predicted or deduced, but now starting from the more fundamental Eq. (11) rather than from Eq. (15).

One of the magneto-optical phenomena of current interest is the inverse Faraday effect (IFE). Note that IFE refers to the induction of magnetization by a circularly polarized incident EM wave in a material[7,14] where the induced magnetization is proportional to the waves' intensity. The IFE is experiencing a resurgence of interest because of its recent observation in magnetically-ordered materials (namely, ferrimagnets and ferromagnets)[8,15–19] and its possible role in optically induced ultrafast magnetization dynamics and reversal (see, e.g., the review of Ref. 9). The origin of the IFE is presently not well understood on a fundamental level (see, e.g., Refs. 20–25). It has recently been represented as an induced optomagnetic field in studies of ultrafast laser-induced magnetization reversal in ferrimagnetic alloys[26].

Here we investigate if the derived AME Hamiltonian can contribute to the IFE effect in the case of a propagating plane EM wave, which thus has only a transverse component to the electric field,

$$\boldsymbol{E}_{\rm ext}(\boldsymbol{r}, t) = -\frac{\partial \boldsymbol{A}}{\partial t} = \mathcal{R}\big(\boldsymbol{E}_0\, e^{i(\boldsymbol{k}\cdot\boldsymbol{r} - \omega t)}\big) \tag{16}$$

where ω is the angular frequency and $\boldsymbol{k}$ is the wavevector. For a general elliptically polarized EM wave propagating along an arbitrary $\boldsymbol{e}_z$ axis,

$$\boldsymbol{E}_0 = \frac{E_0}{\sqrt{2}} \left(\boldsymbol{e}_x + e^{i\eta} \boldsymbol{e}_y\right), \tag{17}$$

where η determines the ellipticity. The vector potential writes according to Eq. (16)

$$\boldsymbol{A}(\boldsymbol{r}, t) = \mathcal{R}\Big(-i \frac{\boldsymbol{E}_0}{\omega} e^{i(\boldsymbol{k}\cdot\boldsymbol{r} - \omega t)}\Big), \tag{18}$$

which, after inserting into the AME coupling Hamiltonian (Eq. (11)) yields:

$$\mathcal{H}^{\rm ind}_{\rm AME} = -\frac{a}{2\omega} \mathcal{R}[\, i\, (\boldsymbol{E}_0 \times \boldsymbol{E}_0^*)] \cdot \boldsymbol{\sigma}. \tag{19}$$

This is precisely the form that has been phenomenologically[7,14] introduced to explain the IFE. In other words, the AME Hamiltonian naturally leads to a straightforward derivation of the IFE effect. Moreover, the Hamiltonian $\mathcal{H}^{\rm ind}_{\rm AME}$ can be rewritten as $-\boldsymbol{B}_{\rm opt} \cdot g\mu_B \boldsymbol{\sigma}$, with g and μ_B being the Landé factor and the Bohr magneton, respectively, and $\boldsymbol{B}_{\rm opt}$ being an optically induced magnetic field given by

$$\begin{aligned}\boldsymbol{B}_{\rm opt} &= \frac{a}{2g\mu_B\omega} \mathcal{R}\left(i\boldsymbol{E}_0 \times \boldsymbol{E}_0^*\right) \\ &= (1+\gamma) \frac{e^2\hbar}{4m^2c^3 g\mu_B\epsilon_0\omega} \sin\eta\, I \boldsymbol{e}_z, \end{aligned} \tag{20}$$

with $I = c\,\epsilon_0 E_0^2/2$ being the intensity of the incoming light. The optomagnetic field $\boldsymbol{B}_{\rm opt}$ is maximal for circularly polarized light, $\eta = \pm\pi/2$, and vanishes for linearly polarized light, $\eta = 0$ or π, as expected from the IFE. In order to estimate the strength of the AME-induced magnetic field, we consider circularly polarized light with an intensity $I = 10$ GW/cm^2, which is large but reachable with contemporary sources, and light of 800 nm wavelength. In the limit $\gamma \longrightarrow 0$, i.e., no screening response of the solid to the applied EM field, the optically induced magnetic field is of the order of 8 μT. On the other hand, the AME coupling has been shown to reproduce the anomalous Hall effect[4]. Assuming that the anomalous Hall effect is fully described by the AME Hamiltonian (11) allows to estimate an upper limit for γ at zero frequency[27], which would correspond to an induced magnetic field in iron as high as 1.5 T.

It is important to realize that Eq. (20) provides the relativistic spin–EM angular momentum-coupling contribution of the IFE (because our starting point is the AME Hamiltonian) and that there is also a part of the inverse Faraday effect that exists even in the absence of spin-orbit coupling[20,22,25].

In summary, the present work derives the existence of the AME Hamiltonian starting from the Dirac equation. Such Hamiltonian is further proven to be related, in some cases and via its expectation value, to the recently proposed coupling between total angular momentum density of the EM field and the magnetization[1] – which has been shown to reproduce or even predict various spin-orbit-driven magneto-electric effects[1,4–6]. Finally, we also demonstrate that this AME Hamiltonian can easily be used to tackle other effects, such as, e.g., the spin–angular momentum-driven contribution to the IFE. We envision that the presently discovered AME coupling Hamiltonian between the electron spin and the angular momentum of an EM wave may lead to the prediction of novel optomagnetic effects effects of fundamental and technological importance.

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