Predicted Coupling of the Electromagnetic Angular Momentum Density with Magnetic Moments

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Coupling of the Electromagnetic Angular Momentum Density with Magnetic Moments: Proof and Consequences

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Abstract

Analytical derivations are developed to demonstrate that (i) the angular moment density associated with an electromagnetic field can directly couple with magnetic moments to produce a physical energy; (ii) this direct coupling explains known, subtle phenomena, including some recently predicted in magnetoelectric materials; and (iii) this coupling also results in novel effects, such as the occurrence of a magnetic anisotropy that is driven by antiferroelectricity.

The angular moment density associated with an electromagnetic field is defined as [1]:

$$\mathcal{J} = \frac{1}{c^2}\mathbf{r} \times (\mathbf{E} \times \mathbf{H}) \tag{1}$$

where $\mathbf{r}$ is the position vector, $\mathbf{E}$ is the electric field, $\mathbf{H}$ is the magnetic field, and c is the speed of light. Interestingly, it was first predicted and then experimentally demonstrated that the angular momentum resulting from the integration of $\mathcal{J}$ over a volume can be *transferred* to microscopic objects, causing these latter to rotate or spin [2–4]. This transfer occurs because of the conservation of angular momentum between the electromagnetic field and the object, and has been put to use to design original devices such as optical tweezers [5] or spanners [6, 7].

Bearing in mind the spin-orbit effect that couples the angular momentum of a particle with its spin [8], it is legitimate to wonder if the angular moment density associated with an electromagnetic field can *directly couple* with magnetic moments, and therefore produce a physical energy. To our surprise, we are not aware that this fundamental question has ever been resolved or even addressed in the literature! As a result, it is currently unknown if the direct coupling between the electromagnetic angular moment density and magnetic moments can exist, and, if it does, what are the physical consequences of such coupling.

The goal of this Letter is to resolve these questions. In particular, we analytically prove, by studying a specific case involving magnetic vortices, that such direct coupling can indeed occur. We then demonstrate that this coupling is at the heart of the recently proposed and subtle spin-current model [9] in magnetoelectric materials (for which magnetic properties can be controlled by electric fields, or conversely, electric properties can be varied by magnetic fields [10]). Moreover, the direct coupling between the electromagnetic angular moment density and magnetic moments also allows for the prediction of novel energy terms that can result in new physical effects. An example of such effects is the occurrence of an *antiferroelectricity-driven* magnetic anisotropy.

Let us first demonstrate that the angular momentum density can directly couple with magnetic moments. For that, we start with the definition of the so-called magnetic toroidal moment, $\mathbf{T}$ [11, 12]:

$$\mathbf{T} = \frac{1}{2}\int (\mathbf{r} \times \mathcal{M})\, d^3r \quad , \tag{2}$$

where $\mathbf{r}$ is the position vector and $\mathcal{M}(\mathbf{r})$ is the magnetization field (which has the units of a magnetic moment per unit volume).

The magnetic toroidal moment is an important physical quantity, since it is, e.g., the order parameter associated with magnetic vortices (see, e.g., Refs. [13, 14] and references therein). Moreover, it is known [10–12, 15] that this toroidal moment can directly couple with the cross product between the electric field and the magnetic field. There is therefore an energy of the form:

$$\mathcal{E} = a\mathbf{T} \cdot (\mathbf{E} \times \mathbf{H}) \quad , \tag{3}$$

where a is a constant. The existence of such energetic term allows, e.g., magnetic vortices to be manipulated and controlled. For instance, the sense of rotation of the magnetic vortices (i.e., clockwise *versus* counterclockwise) can be switched by changing the direction of $\mathbf{E} \times \mathbf{H}$ – similar to the fact that an electric polarization can be switched by an electric field in ferroelectric materials [16].

Inserting Eq. (2) into Eq. (3) for homogeneous $\mathbf{E}$ and $\mathbf{H}$, and using associative properties of the mixed product, we get:

$$\mathcal{E} = -\frac{a}{2} \int (\mathbf{r} \times (\mathbf{E} \times \mathbf{H})) \cdot \mathcal{M}(\mathbf{r}) \; d^3r \tag{4}$$

Equation (4) can be re-written by using the definition of the angular moment density (see Eq. (1)) as:

$$\mathcal{E} = -\frac{ac^2}{2} \int \mathcal{J} \cdot \mathcal{M} \; d^3r \quad , \tag{5}$$

The existence of a dot product between $\mathcal{J}$ and $\mathcal{M}$ on the right side of this latter Equation and the fact that the physical quantity on the left-side of Eq. (5) is an energy demonstrate that the electromagnetic angular momentum density can indeed directly couple with magnetic moments to provide an energy.

Having proved such important result, let us now use it to illustrate an example of a physical effect that such coupling can induce. More precisely, what we have in mind here is to consider a (multiferroic) material possessing both magnetic and electric dipoles around each lattice site, and to reveal that the aforementioned coupling is at the heart of the so-called and recently proposed spin-current model [9]. Let us denote as $\mathbf{d_i}$ the electric dipole existing on the lattice site i, and $\mathbf{m_i}$ and $\mathbf{m_j}$ the magnetic moments centered around the lattice sites i and j, respectively. Following Eq. (1), one can define an angular momentum density produced by site i and acting around site j as:

$$\mathcal{J} = \frac{1}{c^2} \mathbf{r_{i \to j}} \times (\mathbf{E_{i \to j}} \times \mathbf{H_{i \to j}}) \quad , \tag{6}$$

where $\mathbf{E_{i\to j}}$ and $\mathbf{H_{i\to j}}$ are the electric field and magnetic field produced by site i and acting around site j, respectively. $\mathbf{r_{i\to j}}$ is the vector joining the site i to any position contained in the (ionic) volume, V, centered around the site j. For simplicity, we assume that such volume is a sphere of radius R_{ion} (see Fig. 1). We can thus write:

$$\mathbf{r_{i\to j}} = \mathbf{R_{i\to j}} + \delta\mathbf{r} \tag{7}$$

where $\mathbf{R_{i\to j}}$ is the vector joining site i to site j and $\delta\mathbf{r}$ is the vector joining site j to the tip of $\mathbf{r_{i\to j}}$. Since, within the volume V, one can always find two points with opposite $\delta\mathbf{r}$, the integration of $\mathbf{r_{i\to j}}$ around V gives:

$$\int_V \mathbf{r_{i\to j}}\ d^3r = \int_V \mathbf{R_{i\to j}}\ d^3r + \int_V \delta\mathbf{r}\ d^3r = \frac{4\pi R_{ion}^3}{3}\mathbf{R_{i\to j}} \tag{8}$$

since $\mathbf{R_{i\to j}}$ is a constant, $\int_V d^3r = \frac{4\pi R_{ion}^3}{3}$ and $\int_V \delta\mathbf{r}\ d^3r = \mathbf{0}$.

Let us also assume that $\delta\mathbf{r}$ is much smaller in magnitude than $\mathbf{R_{i\to j}}$ (which is usually justified since distances between ions are typically much larger than ionic radius). As a result, $\mathbf{E_{i\to j}}$ and $\mathbf{H_{i\to j}}$ can be taken as constant at any point located inside the volume V and are equal to [1]:

$$\begin{aligned}
\mathbf{E_{i\to j}} &= \frac{1}{4\pi\epsilon_\infty R_{i\to j}^3}\left[3\left(\mathbf{d_i}\cdot\mathbf{e_{i\to j}}\right)\mathbf{e_{i\to j}} - \mathbf{d_i}\right] \\
\mathbf{H_{i\to j}} &= \frac{1}{4\pi R_{i\to j}^3}\left[3\left(\mathbf{m_i}\cdot\mathbf{e_{i\to j}}\right)\mathbf{e_{i\to j}} - \mathbf{m_i}\right]
\end{aligned} \tag{9}$$

where ϵ_∞ is the electronic dielectric constant, and $\mathbf{e_{i\to j}}$ is the unit vector along $\mathbf{R_{i\to j}}$.

Combining Eqs. (6) and Eq. (9) gives:

$$\mathcal{J} = \frac{1}{16\pi^2c^2\epsilon_\infty R_{i\to j}^6}\mathbf{r_{i\to j}} \times \left\{\left(\mathbf{d_i}\times\mathbf{m_i}\right) - 3\mathbf{e_{i\to j}} \times \left[\left(\mathbf{d_i}\cdot\mathbf{e_{i\to j}}\right)\mathbf{m_i} - \left(\mathbf{m_i}\cdot\mathbf{e_{i\to j}}\right)\mathbf{d_i}\right]\right\} \tag{10}$$

According to Eq. (5), the coupling between this angular momentum density and the magnetic moment $\mathbf{m_j}$ inside the volume V centered around site j results in the following energy:

$$\mathcal{E}_{i\to j} = -\frac{ac^2}{2}\frac{3}{4\pi R_{ion}^3}\left(\int_V \mathcal{J}\cdot\mathbf{m_j}\ d^3r\right) \quad , \tag{11}$$

As detailed in the supplemental material, inserting Eq. (10) into Eq. (11), realizing that $\mathbf{m_j}$ can be taken as a constant inside V, and using Eq. (8), as well as properties associated with cross and mixed products, we get:

$$\begin{aligned}
\mathcal{E}_{i\to j} &= \mathcal{E}_{i\to j,1} + \mathcal{E}_{i\to j,2} + \mathcal{E}_{i\to j,3} \quad \textit{with} \\
\mathcal{E}_{i\to j,1} &= -\frac{a}{16\pi^2\epsilon_\infty R^5_{i\to j}} \left(\mathbf{d_i} \times \mathbf{e_{i\to j}}\right) \cdot \left(\mathbf{m_i} \times \mathbf{m_j}\right) \quad , \\
\mathcal{E}_{i\to j,2} &= +\frac{a}{16\pi^2\epsilon_\infty R^5_{i\to j}} \left(\mathbf{d_i} \cdot \mathbf{m_i}\right) \left(\mathbf{e_{i\to j}} \cdot \mathbf{m_j}\right) \quad , \\
\mathcal{E}_{i\to j,3} &= -\frac{a}{16\pi^2\epsilon_\infty R^5_{i\to j}} \left(\mathbf{d_i} \cdot \mathbf{e_{i\to j}}\right) \left(\mathbf{m_i} \cdot \mathbf{m_j}\right)
\end{aligned} \tag{12}$$

Such energy terms characterize the effect of site i on the magnetic moment at site j, and similar expressions can be derived when considering the (reverse) effect of site j on the magnetic moment at site i. Combining these two effects therefore gives:

$$\begin{aligned}
\mathcal{E}_{ij} = \frac{1}{2}\left(\mathcal{E}_{i\to j} + \mathcal{E}_{j\to i}\right) &= -\frac{a}{32\pi^2\epsilon_\infty R^5_{i\to j}} \left([\mathbf{d_i} + \mathbf{d_j}] \times \mathbf{e_{i\to j}}\right) \cdot \left(\mathbf{m_i} \times \mathbf{m_j}\right) \\
&+ \frac{a}{32\pi^2\epsilon_\infty R^5_{i\to j}} \left[\left(\mathbf{d_i} \cdot \mathbf{m_i}\right)\left(\mathbf{e_{i\to j}} \cdot \mathbf{m_j}\right) - \left(\mathbf{d_j} \cdot \mathbf{m_j}\right)\left(\mathbf{e_{i\to j}} \cdot \mathbf{m_i}\right)\right] \\
&- \frac{a}{32\pi^2\epsilon_\infty R^5_{i\to j}} \left([\mathbf{d_i} - \mathbf{d_j}] \cdot \mathbf{e_{i\to j}}\right) \left(\mathbf{m_i} \cdot \mathbf{m_j}\right)
\end{aligned} \tag{13}$$

Let us now consider the case for which the electric dipoles moments are *homogeneous*, that is $\mathbf{d_i} = \mathbf{d_j}$. In that case, it is trivial to show that the third term of Eq. (13) vanishes and that the second term is minus half the first term, which therefore leads to the following energy:

$$\mathcal{E}_{ij} = +b\left(\mathbf{d_i} \times \mathbf{e_{i\to j}}\right) \cdot \left(\mathbf{m_i} \times \mathbf{m_j}\right) \quad , \tag{14}$$

where b is a coefficient equal to $\frac{-a}{32\pi^2\epsilon_\infty R^5_{i\to j}}$.

Remarkably, Eq. (13) characterizes the so-called spin-current model [9, 17], which is a novel magnetoelectric effect that has been recently proposed to explain why a spiral spin structure can generate an electric polarization [9] or how the existence of an electrical polarization can lead to a magnetic cycloid [17] in multiferroics. In other words, our (straightforward) derivations demonstrate that the direct coupling between the electromagnetic angular momentum density and magnetic moments can be thought as being the origin of the "mysterious" spin-current model [18].

Interestingly, the (general) Eq. (13) can also reveal additional novel magneto-electric equations and effects that have never been mentioned in the literature! For instance, let us consider the *antiferroelectric* case for which $\mathbf{d_i} = -\mathbf{d_j}$, with $\mathbf{d_i}$ and $\mathbf{e_{i\to j}}$ perpendicular to each other. Let us also assume that $\mathbf{m_i}$ and $\mathbf{m_j}$ have the same magnitude and both belong to the plane spanned by $\mathbf{d_i}$ and $\mathbf{e_{i\to j}}$. In that case, the first and third terms of Eq. (13) vanish, and it is easy to prove (by using equalities from trigonometry) that one has:

$$\mathcal{E}_{ij} \;=\; \frac{a|\mathbf{d_i}|m^2}{32\pi^2\epsilon_\infty R_{i\to j}^5}\sin(\alpha+\beta) \tag{15}$$

where α is the angle between $\mathbf{d_i}$ and $\mathbf{m_i}$, and β is the angle between $\mathbf{d_i}$ and $\mathbf{m_j}$. This novel energy term therefore desires (through its minization) the sum of α and β to be 90 degrees (270 degrees) if a is negative (positive). As a result, a collinear solution for a *ferromagnetic* material will be to have $\mathbf{m_i}$ and $\mathbf{m_j}$ both making an angle of 45 degrees with respect to $\mathbf{d_i}$, if a is negative. Similarly, a collinear solution for an *antiferromagnetic* material will be to have $\mathbf{m_i}$ and $\mathbf{m_j}$ being antiparallel and $\mathbf{m_i}$ making an angle of 45 degrees with respect to $\mathbf{d_i}$, if a is positive. A a result, Eq. (15) should influence the direction of the easy axis in ferromagnetic and antiferroelectric materials, and will also affect the preferred direction of the antiferromagnetic vector in antiferromagnetic and antiferroelectric materials. In other words, Eq. (15) can be seen as characterizing an *antiferroelectricity-driven magnetic anisotropy*, which is a novel effect to the best of our knowledge.

In summary, this Letter first proves that the electromagnetic angular momentum density can directly couple with magnetic moments (this proof was done here by considering the magnetic toroidal moment and its interaction with the cross-product of the electric field and magnetic field). Secondly, we also demonstrate an important consequence of such coupling, namely the existence of the so-called spin-current model in multiferroics [9]. Thirdly, we show that this direct coupling also leads to the prediction of novel magnetoelectric features (e.g., an antiferroelectricity-driven magnetic anisotropy). Moreover, the Supplementary Material also reveals (in a simple and straightforward manner) that this coupling can originate from spin-orbit and relativistic effects, and can involve a striking product between spin-orbit interactions and electric potential – that also naturally arises from perturbation theory at the second order. Because electromagnetism is fundamental to many branches of physics, chemistry and engineering, it is likely that this direct coupling will explain other subtle effects or will be useful in discovering other novel phenomena in diverse fields of research such as optics, condensed matter, material science, and device physics. We therefore expect that this work would be of great interest and benefit to the scientific community at large.

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[18] Note that the pioneering work of Ref. [9] derived the spin-current model by combining several effects, namely a (i) microscopic electronic Hamiltonian, (ii) a three atom model, (iii) a hole picture (in which oxygen orbitals are empty), (iv) second-order perturbation theory and (v)

double-exchange interactions [19, 20] or superexchange [21] interactions. We humbly believe that our method is more straightforward, more general, and provides a clear picture of the origin of the spin-current model (namely, the direct coupling between the electromagnetic angular momentum density and magnetic moments).

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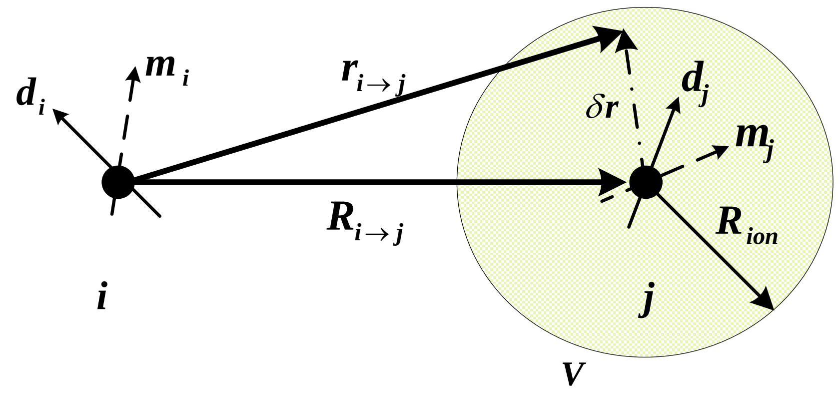
Figure Captions.

FIG. 1. (color online) Schematic of the quantities involved in the derivation of the formula associated with the spin-current model (see Eq. (13) in the text).



m_i
d_i
$r_{i\to j}$
δr
d_j
m_j
$R_{i\to j}$
R_{ion}
i
j
V