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Dynamics of the relativistic electron spin in an electromagnetic field

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Abstract

A relativistic spin operator cannot be uniquely defined within relativistic quantum mechanics. Previously, different proper relativistic spin operators have been proposed, such as spin operators of the Foldy–Wouthuysen and Pryce type, that both commute with the free-particle Dirac Hamiltonian and represent constants of motion. Here we consider the dynamics of a relativistic electron spin in an external electromagnetic field. We use two different Hamiltonians to derive the corresponding spin dynamics. These two are: (a) the Dirac Hamiltonian in the presence of an external field, and (b) the semirelativistic expansion of the same. Considering the Foldy–Wouthuysen and Pryce spin operators we show that these lead to different spin dynamics in an external electromagnetic field, which offers possibilities to distinguish their action. We find that the dynamics of both spin operators involve spin-dependent and spin-independent terms, however, the Foldy–Wouthuysen spin dynamics additionally accounts for the relativistic particle-antiparticle coupling. We conclude that the Pryce spin operator provides a suitable description of the relativistic spin dynamics in a weak-to-intermediate external field, whereas the Foldy–Wouthuysen spin operator is more suitable in the strong field regime.

Keywords: relativistic spin operator, spin dynamics, spin-orbit coupling, strong field physics

1. Introduction

Spin, in quantum mechanics, is an intrinsic property of an elemental particle e.g., of the electron. However, in contrast to nonrelativistic quantum mechanics, the definition of the spin operator is not unique in relativistic quantum mechanics [1–5]. In nonrelativistic quantum mechanics, the spin is expressed by the Pauli spin matrices as $\boldsymbol{\sigma}$ and the corresponding spin angular momentum by $\boldsymbol{S} = \frac{\boldsymbol{\sigma}}{2}$ (assuming units such that $\hbar = 1$). The latter definition is valid for the two-component Schrödinger or Pauli Hamiltonian that relates directly the spin operator to the Pauli spin matrices. However, in a relativistic formulation the spin angular momentum cannot be defined separately because the total angular momentum has to be conserved. Therefore, the definition of spin angular momentum depends on the definition of the orbital angular momentum. Generally, the orbital angular momentum is defined as $\boldsymbol{L} = \boldsymbol{r} \times \boldsymbol{p}$ such that the total angular momentum is calculated as $\boldsymbol{J} = \boldsymbol{L} + \boldsymbol{S}$ in a nonrelativistic framework. Even so, in relativistic quantum mechanics, the position operator is not uniquely defined and, consequently, the spin angular momentum does not have a unique definition [1, 2, 6]. In fact, for both the orbital and spin angular momentum several definitions have been proposed [6].

While the definition of the relativistic spin operator might seem a semantic issue, its formulation does in fact matter when relativistic spin dynamics is considered. Spin dynamics has previously been computed starting from the nonrelativistic spin operator, i.e. the Pauli spin matrices $\boldsymbol{\sigma}$ [7, 8]. The resulting equation of motion is found to be composed of spin precession, spin relaxation and even spin nutation (inertial dynamics), terms that are consistent with the well-known Landau–Lifshitz–Gilbert (LLG) equation of spin dynamics. Even

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though the LLG equation has been used for the spin dynamics at ultrashort timescales, its applicability at these timescales has been questioned [9]. However, the relativistic spin dynamics has not yet been derived from a relativistic spin operator. With this objective, we derive in this article the spin dynamics for relativistic spin operators, in particular, we treat the previously proposed proper relativistic spin operators due to Foldy and Wouthuysen [10, 11] and Pryce [12, 13]. We consider three different Hamiltonians to derive the relativistic spin dynamics: (1) the free-particle Dirac Hamiltonian, (2) the Dirac Hamiltonian in an electromagnetic (EM) environment, and (3) the Foldy–Wouthuysen (FW) transformation of the Dirac Hamiltonian in an EM environment. The results show that the corresponding spin dynamics leads to the LLG equation of motion, however with additional contributions due to the relativistic spin operator formulations. Comparing the relativistic dynamics for an electron spin in an EM field, we draw the conclusion that the Pryce spin operator provides a suitable formulation of the relativistic electron spin dynamics in the weak to intermediate field regime, however, the FW spin operator is more applicable for describing spin in the relativistic strong field regime.

In the following we first introduce the relativistic spin operators, especially the FW and Pryce spin operators. Thereafter, in section 3 we formulate the three different Hamiltonians that will be used to evaluate the relativistic spin dynamics. Then, in section 4 we derive the spin dynamics corresponding to the FW and Pryce spin operators and discuss the obtained results. Conclusions are drawn in section 5.

2. Relativistic spin operators

The free-particle Dirac Hamiltonian reads [14–16]

$$\mathcal{H}_{\rm D}^{0} = c\,\boldsymbol{\alpha}\cdot\boldsymbol{p} + \beta m_0 c^2, \tag{1}$$

with the rest mass m_0, and $\boldsymbol{\alpha}$ and β are the 4×4 Dirac matrices which obey the following relations [17]

$$\begin{aligned} \alpha_i^2 = \mathbb{1}, \qquad \beta^2 = \mathbb{1}, \qquad \alpha_i\alpha_j + \alpha_j\alpha_i = 2\delta_{ij}, \\ \alpha_i\beta + \beta\alpha_i = 0, \end{aligned} \tag{2}$$

where $\mathbb{1}$ is the 4×4 identity matrix. The corresponding spin operator is a four-component operator that describes both the particle spin up (down) and antiparticle spin up (down) states. The Dirac spin operator has hence the definition $\boldsymbol{S}_{\rm D} = \frac{\boldsymbol{\Sigma}}{2}$, with the components of the operator $\boldsymbol{\Sigma}$ $(= \mathbb{1}\otimes\boldsymbol{\sigma})$ as

$$\Sigma_j = -i\alpha_k\alpha_l. \tag{3}$$

In addition, the orbital angular momentum is given as $\boldsymbol{L}_{\rm D} = \boldsymbol{r}\times\boldsymbol{p}$, which has to be multiplied by a 2-units block diagonal matrix of 2×2. The total angular momentum is then given by $\boldsymbol{J}_{\rm D} = \boldsymbol{L}_{\rm D} + \boldsymbol{S}_{\rm D}$ and it recovers the angular momentum in the nonrelativistic framework when taken in two-component form.

The spin does not couple to the orbital angular momentum in nonrelativistic quantum mechanics, in fact, the spin operator $\boldsymbol{S}$ is a constant of motion when the Schrödinger Hamiltonian is considered. However, for a free-particle Dirac Hamiltonian, the calculation of the spin dynamics with the Dirac spin operator reveals

$$\frac{{\rm d}\boldsymbol{S}_{\rm D}}{{\rm d}t} = -c\,\boldsymbol{\alpha}\times\boldsymbol{p}, \tag{4}$$

meaning that the Dirac spin operator is not a constant of motion. As one expects, the corresponding dynamics contains the particle–antiparticle coupling strength, following this feature of the Dirac Hamiltonian. Moreover, the dynamics suggests that the Dirac matrices α_i are coupled to the orbital angular momentum via $\boldsymbol{p} = -i\nabla$. Furthermore, it has been shown that the eigenvalues of the Dirac spin operator deviate from $\pm 1/2$ for materials having higher atomic numbers [18]. The latter is understandable because, for higher atomic numbers, the spin cannot be considered as an independent quantity, rather the spin is coupled to the orbital degrees of freedom due to larger spin–orbit coupling. Thus, the two major drawbacks of the Dirac spin operator are that (a) it does not commute with the free-particle Dirac Hamiltonian, (b) the eigenvalue does not correspond to $\pm 1/2$ for systems with higher atomic number, which implies that the Dirac spin operator cannot be considered as a proper relativistic spin operator.

A proper relativistic spin operator should have the following properties [4, 18]:

(a) It has to commute with the relativistic free-particle Dirac equation. This implies that the spin operator is a constant of motion for a Dirac free-particle.
(b) It has to obey the SU(2) algebra of spin operators. The commutator of two spin operators should follow the relation
$$\left[S_i, S_j\right] = i\epsilon_{ijk}S_k, \tag{5}$$
where ϵ_{ijk} is the anti-symmetric Levi–Civita tensor.
(c) The spin operator must have two eigenvalues of $\pm\frac{1}{2}$.
(d) The total angular momentum has to be conserved.

There have been a number of relativistic spin operators reported in the literature [4, 5, 18–21]. However, all of these existing spin operators do not obey all the aforementioned conditions. It is known that *only* the relativistic Foldy–Wouthuysen and Pryce spin operators [10–13] satisfy all the above-mentioned properties. Therefore, one can infer that they can be considered as proper spin operators [18].

In the following, we calculate the spin dynamics corresponding to both of these operators for a system excited by an electromagnetic field (e.g., a laser pulse).

2.1. FW spin operator

The FW spin operator has the following definition [10, 11, 17, 22, 23]

$$\boldsymbol{S}_{\rm FW} = \frac{1}{2}\boldsymbol{\Sigma} + \frac{i\beta\boldsymbol{p}\times\boldsymbol{\alpha}}{2E_p} - \frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{2E_p(E_p+m_0c^2)}, \tag{6}$$

with the energy $E_p = \sqrt{p^2c^2 + m_0^2c^4}$. Correspondingly, the position operator is also defined as

$$\boldsymbol{r}_{\mathrm{FW}} = \boldsymbol{r} + \frac{i\beta\boldsymbol{\alpha}}{2E_p} - \frac{i\beta\left(\boldsymbol{\alpha}\cdot\boldsymbol{p}\right)\boldsymbol{p} + (\boldsymbol{\Sigma}\times\boldsymbol{p})\,|\boldsymbol{p}|}{2E_p(E_p+m_0c^2)|\boldsymbol{p}|}, \tag{7}$$

such that the total angular momentum is exactly the same as that of the nonrelativistic case i.e., $\boldsymbol{J}_{\mathrm{FW}} = \boldsymbol{L}_{\mathrm{FW}} + \boldsymbol{S}_{\mathrm{FW}} = \boldsymbol{r}_{\mathrm{FW}}\times\boldsymbol{p} + \boldsymbol{S}_{\mathrm{FW}} = \boldsymbol{r}\times\boldsymbol{p} + \frac{\boldsymbol{\Sigma}}{2}$. This construction 'made by hand' reflects that the total angular momentum has to be conserved, and has to be equal to the total angular momentum for the Pauli representation when we consider the two-component form. In the following, we provide detailed derivations that show that the FW spin operator follows the above-mentioned properties of a proper spin operator:

(a) The commutation algebra allows us to calculate the time derivative as

$$\begin{aligned}\frac{\mathrm{d}\boldsymbol{S}_{\mathrm{FW}}}{\mathrm{d}t} &= \frac{1}{i}\left[\boldsymbol{S}_{\mathrm{FW}}, \mathcal{H}^0_{\mathrm{D}}\right] \\ &= \frac{1}{2i}\left[\boldsymbol{\Sigma}, c\,\boldsymbol{\alpha}\cdot\boldsymbol{p}\right] + \frac{1}{2E_p}\left[\beta\boldsymbol{p}\times\boldsymbol{\alpha}, c\,\boldsymbol{\alpha}\cdot\boldsymbol{p}\right] \\ &\quad - \frac{1}{2i}\left[\frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{E_p(E_p+m_0c^2)}, c\,\boldsymbol{\alpha}\cdot\boldsymbol{p}\right] \\ &= 0.\end{aligned} \tag{8}$$

While deriving the commutators, we have used the following results: $[\boldsymbol{\Sigma}, \boldsymbol{\alpha}\cdot\boldsymbol{p}] = -2i\boldsymbol{\alpha}\times\boldsymbol{p}$ and $[\beta\boldsymbol{p}\times\boldsymbol{\alpha}, \boldsymbol{\alpha}\cdot\boldsymbol{p}] = \beta\{\boldsymbol{p}\times\boldsymbol{\alpha}, \boldsymbol{\alpha}\cdot\boldsymbol{p}\}_+$ which vanishes after some algebra. However, the third commutator can be calculated as: $[\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p}), \boldsymbol{\alpha}\cdot\boldsymbol{p}] = -2ip^2\boldsymbol{\alpha}\times\boldsymbol{p}$ and it cancels with the result from first commutator. Equation (8) states that the FW spin operator is a constant of motion when the free-particle Dirac Hamiltonian is considered. This result is expected according to the first condition of a proper spin operator [18].

(b) We calculate the spin angular momentum commutator relations as

$$\begin{aligned}\left[S^i_{\mathrm{FW}}, S^j_{\mathrm{FW}}\right] &= \frac{1}{2}\left[\boldsymbol{\Sigma}^i, \frac{1}{2}\boldsymbol{\Sigma}^j + \frac{i\beta\boldsymbol{p}\times\boldsymbol{\alpha}}{2E_p}\right|^j \\ &\quad \left. - \frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{2E_p(E_p+m_0c^2)}\right|^j\Bigg] \\ &\quad + \frac{i}{2E_p}\left[\beta\boldsymbol{p}\times\boldsymbol{\alpha}\Big|^i, \frac{1}{2}\boldsymbol{\Sigma}^j + \frac{i\beta\boldsymbol{p}\times\boldsymbol{\alpha}}{2E_p}\right|^j \\ &\quad \left. - \frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{2E_p(E_p+m_0c^2)}\right|^j\Bigg] \\ &\quad - \left[\frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{2E_p(E_p+m_0c^2)}\Big|^i, \frac{1}{2}\boldsymbol{\Sigma}^j + \frac{i\beta\boldsymbol{p}\times\boldsymbol{\alpha}}{2E_p}\right|^j \\ &\quad \left. - \frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{2E_p(E_p+m_0c^2)}\right|^j\Bigg].\end{aligned} \tag{9}$$

Evidently, all the commutators in the first line and the first commutators in the second and third line of equation (9) contribute. We calculate the contributing commutators as:

$$\frac{1}{2}\left[\boldsymbol{\Sigma}^i, \frac{1}{2}\boldsymbol{\Sigma}^j\right] = i\epsilon_{ijk}\frac{\boldsymbol{\Sigma}^k}{2}, \tag{10}$$

$$\begin{aligned}&\left[\frac{1}{2}\boldsymbol{\Sigma}^i, \frac{i}{2E_p}\beta\boldsymbol{p}\times\boldsymbol{\alpha}\Big|^j\right] + \left[\frac{i}{2E_p}\beta\boldsymbol{p}\times\boldsymbol{\alpha}\Big|^i, \frac{1}{2}\boldsymbol{\Sigma}^j\right] \\ &\quad = (i\epsilon_{ijk})\frac{i\beta}{2E_p}\boldsymbol{p}\times\boldsymbol{\alpha}\Big|^k,\end{aligned} \tag{11}$$

$$\begin{aligned}&\left[\frac{1}{2}\boldsymbol{\Sigma}^i, \frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{2E_p(E_p+m_0c^2)}\Big|^j\right] + \left[\frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{2E_p(E_p+m_0c^2)}\Big|^i, \frac{1}{2}\boldsymbol{\Sigma}^j\right] \\ &\quad = -(i\epsilon_{ijk})\frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{2E_p(E_p+m_0c^2)}\Big|^k.\end{aligned} \tag{12}$$

Therefore, the commutation relation of the FW spin operators can be recast as

$$\begin{aligned}\left[S^i_{\mathrm{FW}}, S^j_{\mathrm{FW}}\right] &= i\epsilon_{ijk}\left[\frac{\boldsymbol{\Sigma}^k}{2} + \frac{i\beta}{2E_p}\boldsymbol{p}\times\boldsymbol{\alpha}\right|^k \\ &\quad \left. - \frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{2E_p(E_p+m_0c^2)}\right|^k\Bigg] = i\epsilon_{ijk}S^k_{\mathrm{FW}}.\end{aligned} \tag{13}$$

(c) The eigenvalues of the FW spin operator have been computed from the expectation values, using the hydrogenic ground state in reference [18]. If the FW spin operator along the z-direction is represented by S^z_{FW} and the hydrogenic ground state degenerate wave functions by $\Psi_\uparrow$ and $\Psi_\downarrow$, the calculation of the expectation values shows that $\langle\Psi_\uparrow|S^z_{\mathrm{FW}}|\Psi_\uparrow\rangle = +\frac{1}{2}$ (see figure 1 in reference [18]) even for elements having higher atomic number.

(d) Using the FW spin and position operators, one can calculate the total angular momentum as following,

$$\begin{aligned}\boldsymbol{J}_{\mathrm{FW}} &= \boldsymbol{r}_{\mathrm{FW}}\times\boldsymbol{p} + \boldsymbol{S}_{\mathrm{FW}} \\ &= \boldsymbol{r}\times\boldsymbol{p} + \frac{i\beta\boldsymbol{\alpha}\times\boldsymbol{p}}{2E_p} \\ &\quad - \frac{i\beta\left(\boldsymbol{\alpha}\cdot\boldsymbol{p}\right)\boldsymbol{p}\times\boldsymbol{p} + (\boldsymbol{\Sigma}\times\boldsymbol{p})\times\boldsymbol{p}|\boldsymbol{p}|}{2E_p(E_p+m_0c^2)|\boldsymbol{p}|} \\ &\quad + \frac{1}{2}\boldsymbol{\Sigma} + \frac{i\beta\boldsymbol{p}\times\boldsymbol{\alpha}}{2E_p} - \frac{\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})}{2E_p(E_p+m_0c^2)} \\ &= \boldsymbol{r}\times\boldsymbol{p} + \frac{\boldsymbol{\Sigma}}{2}.\end{aligned} \tag{14}$$

Therefore, the FW total angular momentum is conserved and equals to the total angular momentum for a Dirac spin operator. On account of conditions (a) to (d), the FW spin operator can be taken as a proper relativistic spin operator.

2.2. *Pryce spin operator*

The Pryce spin operator has the following definition [12, 13]:

$$\boldsymbol{S}_{\mathrm{Py}} = \frac{1}{2}\beta\boldsymbol{\Sigma} + \frac{1}{2}\left(1-\beta\right)\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}, \tag{15}$$

and the corresponding position operator has the form

$$\boldsymbol{r}_{\mathrm{Py}} = \boldsymbol{r} - \frac{1}{2}(1-\beta)\frac{\boldsymbol{\Sigma}\times\boldsymbol{p}}{p^2}, \tag{16}$$

such that the total angular momentum is written as $\boldsymbol{J}_{\mathrm{Py}} = \boldsymbol{L}_{\mathrm{Py}} + \boldsymbol{S}_{\mathrm{Py}} = \boldsymbol{r}_{\mathrm{Py}}\times\boldsymbol{p} + \boldsymbol{S}_{\mathrm{Py}} = \boldsymbol{r}\times\boldsymbol{p} + \frac{\boldsymbol{\Sigma}}{2}$. The derived total angular momentum for the Pryce spin and orbital momentum operator is equal to the total angular momentum in the Pauli representation as argued earlier. In the below, we show that the Pryce operator obeys the properties of a proper spin operator.

(a) Starting from free-particle Dirac Hamiltonian, we can calculate the Pryce spin dynamics as follows

$$\begin{aligned}\frac{\mathrm{d}\boldsymbol{S}_{\mathrm{Py}}}{\mathrm{d}t} &= \frac{1}{i}\left[\boldsymbol{S}_{\mathrm{Py}}, \mathcal{H}_{\mathrm{D}}^{0}\right]\\ &= \frac{1}{2i}\left[\beta\boldsymbol{\Sigma}, c\boldsymbol{\alpha}\cdot\boldsymbol{p}\right] + \frac{1}{2i}\left[(1-\beta)\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}, c\boldsymbol{\alpha}\cdot\boldsymbol{p}\right]\\ &= 0.\end{aligned} \tag{17}$$

We have used the following results from the commutator: $[\beta\boldsymbol{\Sigma}, \boldsymbol{\alpha}\cdot\boldsymbol{p}] = \beta\{\boldsymbol{\Sigma}, \boldsymbol{\alpha}\cdot\boldsymbol{p}\}_{+} = \frac{2}{3}(\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha})\boldsymbol{p}$, which is evidently off-diagonal in particle–antiparticle space. The second commutator vanishes i.e., $[(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}, \boldsymbol{\alpha}\cdot\boldsymbol{p}] = 0$, however, the last commutator in equation (17) results $[\beta(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}, \boldsymbol{\alpha}\cdot\boldsymbol{p}] = \beta\{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}, \boldsymbol{\alpha}\cdot\boldsymbol{p}\}_{+} = \frac{2}{3}p^2(\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha})\boldsymbol{p}$, which exactly cancels with the first commutator, giving, as expected, that the Pryce spin operator is a constant of motion for $\mathcal{H}_{\mathrm{D}}^{0}$.

(b) The Pryce spin operator commutation relation follows from

$$\begin{aligned}\left[\boldsymbol{S}_{\mathrm{Py}}^{i}, \boldsymbol{S}_{\mathrm{Py}}^{j}\right] = \frac{1}{2}&\left[\beta\boldsymbol{\Sigma}\Big|^{i}, \frac{1}{2}\beta\boldsymbol{\Sigma}\Big|^{j} + \frac{1}{2}(1-\beta)\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}\Big|^{j}\right]\\ &+\left[\frac{1}{2}(1-\beta)\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}\Big|^{i}, \frac{1}{2}\beta\boldsymbol{\Sigma}\Big|^{j}\right.\\ &\left.+\frac{1}{2}(1-\beta)\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}\Big|^{j}\right].\end{aligned} \tag{18}$$

All the commutators in the first line and the first commutator in the second line of equation (18) will contribute. We calculate each commutator in the below

$$\left[\frac{1}{2}\beta\boldsymbol{\Sigma}\Big|^{i}, \frac{1}{2}\beta\boldsymbol{\Sigma}\Big|^{j}\right] = i\epsilon_{ijk}\beta^2\frac{\boldsymbol{\Sigma}^{k}}{2}, \tag{19}$$

$$\begin{aligned}&\left[\frac{1}{2}\beta\boldsymbol{\Sigma}\Big|^{i}, \frac{(1-\beta)}{2}\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}\Big|^{j}\right] + \left[\frac{(1-\beta)}{2}\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}\Big|^{i}, \frac{1}{2}\beta\boldsymbol{\Sigma}\Big|^{j}\right]\\ &= (i\epsilon_{ijk})\beta(1-\beta)\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{2p^2}\Big|^{k}.\end{aligned} \tag{20}$$

Therefore, combining equations (19) and (20), we get the commutator relation for Pryce operators,

$$\begin{aligned}\left[\boldsymbol{S}_{\mathrm{Py}}^{i}, \boldsymbol{S}_{\mathrm{Py}}^{j}\right] &= i\epsilon_{ijk}\beta\left[\beta\frac{\boldsymbol{\Sigma}^{k}}{2} + (1-\beta)\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{2p^2}\Big|^{k}\right]\\ &= i\epsilon_{ijk}\beta\boldsymbol{S}_{\mathrm{Py}}^{k}.\end{aligned} \tag{21}$$

(c) As discussed for the FW spin operator, the eigenvalues of the Pryce spin operator have been calculated using hydrogenic ground state wave functions as $\langle\Psi_{\uparrow}|S_{\mathrm{Py}}^{z}|\Psi_{\uparrow}\rangle$ in reference [18]. In this case, the eigenvalues are found to be exactly $+\frac{1}{2}$, even for the higher atomic numbers.

(d) Lastly, the total angular momentum for the Pryce spin operator can be calculated as

$$\begin{aligned}\boldsymbol{J}_{\mathrm{Py}} &= \boldsymbol{r}_{\mathrm{Py}}\times\boldsymbol{p} + \boldsymbol{S}_{\mathrm{Py}}\\ &= \boldsymbol{r}\times\boldsymbol{p} - \frac{1}{2}(1-\beta)\frac{(\boldsymbol{\Sigma}\times\boldsymbol{p})\times\boldsymbol{p}}{p^2} + \frac{1}{2}\beta\boldsymbol{\Sigma}\\ &\quad+\frac{1}{2}(1-\beta)\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}\\ &= \boldsymbol{r}\times\boldsymbol{p} - \frac{1}{2}(1-\beta)\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})p - \boldsymbol{\Sigma}p^2}{p^2}\\ &\quad+\frac{1}{2}\beta\boldsymbol{\Sigma} + \frac{1}{2}(1-\beta)\frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}\\ &= \boldsymbol{r}\times\boldsymbol{p} + \frac{\boldsymbol{\Sigma}}{2}.\end{aligned} \tag{22}$$

Therefore, the total angular momentum is conserved for the Pryce spin operator. Consequently, the Pryce spin operator can be considered as a proper spin operator, similar to the FW spin operator.

A striking difference between FW and Pryce spin operators is that the FW spin operator contains a particle–antiparticle coupling term, i.e., the second term of equation (6), however, such coupling terms do not appear in the Pryce spin operator in equation (15). One immediately notices that the spin operators contain *not only* the spin angular momentum, *but also*, the orbital angular momentum in the form of $\boldsymbol{p}$. The same is valid for the position operators as well, because of the following reasons. For the FW and Pryce position operators, we obtain (neglecting higher-order terms)

$$r_{\mathrm{FW}}^2 = r^2 + \frac{1}{4E_p^2} - \frac{i\beta(\boldsymbol{r}\cdot\boldsymbol{p})(\boldsymbol{\alpha}\cdot\boldsymbol{p})}{E_p(E_p+m_0c^2)|\boldsymbol{p}|} - \frac{\boldsymbol{\Sigma}\cdot\boldsymbol{L}}{E_p(E_p+m_0c^2)}, \tag{23}$$

$$r_{\mathrm{Py}}^2 = r^2 + (1-\beta)\frac{\boldsymbol{\Sigma}\cdot\boldsymbol{L}}{p^2}, \tag{24}$$

respectively. Here, the last correction terms represent the well-known spin–orbit coupling that is missing in a nonrelativistic description. Note that there is another relativistic correction term that appears in the FW position operator which is notably off-diagonal in the particle–antiparticle Hilbert space. Having these proper relativistic spin operators, we derive their spin dynamics, particularly, in an applied EM field. While both operators are proper spin operators, their formulation is evidently different, and it is unknown which spin operator provides a more suitable description of the dynamics. In particular, we are keen to understand the effects of relativistic coupling terms within the corresponding spin dynamics.

3. Relativistic Hamiltonians

For deriving the spin dynamics, we consider three different Hamiltonians. The first one is the Dirac free-particle Hamiltonian that has already been introduced in equation (1). The second one is the Dirac equation in the presence of an external EM field that is described by the magnetic vector and scalar potentials as $\boldsymbol{A}(\boldsymbol{r}, t)$ and $\phi(\boldsymbol{r}, t)$. This modified Dirac equation can be expressed by the minimal coupling as [17]

$$\mathcal{H}_{\rm D}^{\rm EM} = c\,\boldsymbol{\alpha}\cdot(\boldsymbol{p}-e\boldsymbol{A}) + \beta m_0 c^2 + e\phi. \tag{25}$$

Note that we have not included magnetic exchange interaction in the following derivation because of its additional complexity. A rigorous calculation of spin dynamics with magnetic exchange for the nonrelativistic spin operator can be found in references [8, 24].

Now, we perform the FW transformation of the above Hamiltonian and transform the Hamiltonian as an even Hamiltonian [10, 17, 23, 25]. The FW transformation can be summarized as $\mathcal{H}_{\rm FW} = {\rm e}^{iU}\left(\mathcal{H}_{\rm D}^{\rm EM} - i\frac{\partial}{\partial t}\right){\rm e}^{-iU} + i\frac{\partial}{\partial t}$, where U defines a unitary operator obtained from the odd terms (i.e., off-diagonal in the particle–antiparticle space) of the Hamiltonian $\mathcal{H}_{\rm D}^{\rm EM}$. The nonrelativistic FW transformed Hamiltonian along with the relativistic corrections of equation (25) takes the form [26, 27]

$$\mathcal{H}_{\rm FW} = \beta m_0 c^2 + \beta\left(\frac{\mathcal{O}^2}{2m_0c^2} - \frac{\mathcal{O}^4}{8m_0^3c^6}\right) + \mathcal{E} - \frac{1}{8m_0^2c^4}\left[\mathcal{O},\left[\mathcal{O},\mathcal{F}\right]\right] + \frac{\beta}{16m_0^3c^6}\left\{\mathcal{O},\left[\left[\mathcal{O},\mathcal{F}\right],\mathcal{F}\right]\right\}, \tag{26}$$

with the following definitions of odd and even terms $\mathcal{O} = c\,\boldsymbol{\alpha}\cdot(\boldsymbol{p}-e\boldsymbol{A})$ and $\mathcal{E} = e\phi$, respectively. $[A, B]$ defines the commutator, while $\{A, B\}$ defines the anti-commutator for any two given operators A and B. Within the FW transformation, the even terms and $i\frac{\partial}{\partial t}$ transform in a similar way, therefore, we introduce a combined term $\mathcal{F} = \mathcal{E} - i\frac{\partial}{\partial t}$ [28–31]. We mention, first, that a *relativistic* FW transformation has been developed in reference [29] and, second, that with a FW transformation always only an approximate FW Hamiltonian can be calculated. We calculate the corresponding four-component diagonalized Hamiltonian in the particle–antiparticle space that has the form [32–34]

$$\begin{aligned}\mathcal{H}_{\rm FW} &= \beta m_0 c^2 + \frac{\beta(\boldsymbol{p}-e\boldsymbol{A})^2}{2m_0} - \frac{e\beta}{2m_0}\boldsymbol{\Sigma}\cdot\boldsymbol{B} - \frac{\beta(\boldsymbol{p}-e\boldsymbol{A})^4}{8m_0^3c^2} \\ &+ \frac{e\beta}{8m_0^3c^2}\left\{(\boldsymbol{p}-e\boldsymbol{A})^2, \boldsymbol{\Sigma}\cdot\boldsymbol{B}\right\} - \frac{\beta e^2B^2}{8m_0^3c^2} - \frac{e}{8m_0^2c^2}\nabla\cdot\boldsymbol{E} \\ &+ \frac{e}{8m_0^2c^2}\boldsymbol{\Sigma}\cdot\left[(\boldsymbol{p}-e\boldsymbol{A})\times\boldsymbol{E} - \boldsymbol{E}\times(\boldsymbol{p}-e\boldsymbol{A})\right] \\ &- \frac{ie\beta}{16m_0^3c^4}\boldsymbol{\Sigma}\cdot\left[(\boldsymbol{p}-e\boldsymbol{A})\times\frac{\partial\boldsymbol{E}}{\partial t} + \frac{\partial\boldsymbol{E}}{\partial t}\times(\boldsymbol{p}-e\boldsymbol{A})\right].\end{aligned} \tag{27}$$

We have used the following definitions for the Maxwell fields: $\boldsymbol{B} = \nabla\times\boldsymbol{A}, \boldsymbol{E} = -\frac{\partial\boldsymbol{A}}{\partial t} - \nabla\phi$. The above-derived Hamiltonian is very crucial for understanding the light-particle (antiparticle) interaction at low energy excitation. Equation (27) can be understood as comprising of nonrelativistic terms and relativistic terms [32]. The first term describes the rest mass energy which has to be subtracted from the total energy in order to obtain the Pauli Hamiltonian for quantum particles. The second term describes the kinetic energy term in the Schrödinger equation. The third term is the direct Zeeman coupling of spins with the external magnetic field. The fourth term is the representation of relativistic mass correction terms. The fifth term is an indirect coupling of spins with the external fields. The sixth term is the relativistic correction to the Zeeman coupling. The seventh term explains the Darwin term. The last two terms represent the generalized form of spin–orbit coupling. We note that the direct coupling terms of the spin and the external field are the important ones to describe the corresponding interactions and dynamics [35, 36], however, the indirect coupling terms could also be interesting as well [36, 37]. We also mention that a full Hamiltonian together with the exchange interaction has also been derived in earlier works where, in addition, the relativistic corrections to the exchange interactions are obtained [8, 24, 32, 38, 39]. The Hamiltonian in equation (27) has been used in calculating the general spin dynamics with the Pauli spin operator [7, 8, 24, 26, 32–35, 40–43]. We comment that the calculated spin dynamics could explain the precession, spin relaxation of Gilbert type and even nutation dynamics of a single spin [34]. However, the derivation of the spin dynamics has been calculated using a two-component extended Pauli Hamiltonian and the nonrelativistic spin operator. Here, our goal is to calculate the spin dynamics from relativistic spin operators.

The spin–orbit coupling terms can be recast in a more simplified form by using the well-known Maxwell's equations. Moreover, we can ignore the rest mass energy and constant energy terms in the Hamiltonian of equation (27), because we work out the dynamical equation of motion. The rest of the terms can be simplified to

$$\begin{aligned}\mathcal{H}'_{\rm FW} &= \frac{\beta(\boldsymbol{p}-e\boldsymbol{A})^2}{2m_0} - \frac{e\beta}{2m_0}\boldsymbol{\Sigma}\cdot\boldsymbol{B} - \frac{\beta(\boldsymbol{p}-e\boldsymbol{A})^4}{8m_0^3c^2} + \frac{\beta}{8m_0^3c^2} \\ &\times\left\{(\boldsymbol{p}-e\boldsymbol{A})^2, \boldsymbol{\Sigma}\cdot\boldsymbol{B}\right\} - \frac{e}{8m_0^2c^2}\nabla\cdot\boldsymbol{E} - \frac{\hbar e}{8m_0^2c^2}\boldsymbol{\Sigma} \\ &\cdot\left[2\boldsymbol{E}(\boldsymbol{p}-e\boldsymbol{A}) - i\hbar\frac{\partial\boldsymbol{B}}{\partial t}\right] + \frac{e\beta}{16m_0^3c^4}\boldsymbol{\Sigma}\cdot\frac{\partial^2\boldsymbol{B}}{\partial t^2}.\end{aligned} \tag{28}$$

Further, we can also ignore the Darwin term in our calculation because the Darwin term involves the density of charges according to the Maxwell theory. As we mentioned earlier, the direct coupling terms provide an opportunity to directly manipulate the spins. Therefore, we evaluate in the following the spin dynamics with those terms.

Traditionally one is interested in the direct spin-EM field coupling terms to derive the spin dynamics [35, 36, 44, 45]. However, the definition of FW or Pryce spin operator suggests that one also needs to consider the terms that do not explicitly depend on the spins. The reason is that the orbital angular momentum enters in the relativistic spin operators in the form of $\boldsymbol{p}$. Now, the derivation of spin dynamics follows the time evolution of spin operators that involves the commutators of spin operators with the considered Hamiltonian terms. The commutators of the nonrelativistic Pauli spin operator with spin-independent terms do not contribute to the dynamics. However, for the relativistic spin operator the spin-independent terms have to be considered as well. Therefore, we restrict our derivations to the following FW transformed Hamiltonian

$$\mathcal{H}^{\rm spin}_{\rm direct} = \frac{\beta(\boldsymbol{p}-e\boldsymbol{A})^2}{2m_0} - \frac{e\beta}{2m_0}\boldsymbol{\Sigma}\cdot\boldsymbol{B} - \frac{e}{8m_0^2c^2} \times \boldsymbol{\Sigma}\cdot\left[2\boldsymbol{E}\times(\boldsymbol{p}-e\boldsymbol{A}) - i\frac{\partial\boldsymbol{B}}{\partial t}\right] + \frac{e\beta}{16m_0^3c^4}\boldsymbol{\Sigma}\cdot\frac{\partial^2\boldsymbol{B}}{\partial t^2}. \tag{29}$$

Note that the other relativistic terms will contribute to the dynamical equation of motion as well, however, for simplicity of the calculations, we consider only the above-mentioned direct spin-field interaction terms, which are expected to constitute the main contribution. Moreover, equation (29) contains the linear and quadratic interactions in the field, $\boldsymbol{A}(\boldsymbol{r},t)$. In the below, we calculate the spin dynamics with the linear-order interaction terms with the gauge choice, $\boldsymbol{A} = \frac{\boldsymbol{B}\times\boldsymbol{r}}{2}$ which holds for uniform (*slowly-varying*) magnetic field such that $\nabla\times\boldsymbol{A} = \boldsymbol{B}$ and $\nabla\cdot\boldsymbol{A} = 0$.

4. Derivation of spin dynamics

4.1. FW spin operator

In the presence of an external electromagnetic field, the form of the FW spin operator can be obtained using the unitary operator U. However, the choice of the unitary operator (along with the external field) is non-trivial and cumbersome. Therefore, we use the FW spin operator of equation (6), however, with the replacement of $\boldsymbol{p}\to(\boldsymbol{p}-e\boldsymbol{A})$ [4, 46]. Note that, by doing so, the FW spin operator becomes field dependent [47, 48]. To derive the spin dynamics we calculate the Heisenberg operator dynamics [25].

4.1.1. Dirac Hamiltonian with EM field. The spin dynamics for the FW spin operator for the Dirac equation with an external EM field can be calculated as follows,

$$\begin{aligned}
\frac{\mathrm{d}\boldsymbol{S}_{\rm FW}}{\mathrm{d}t} &= \frac{1}{i}\left[\boldsymbol{S}_{\rm FW}, \mathcal{H}^{\rm EM}_{\rm D}\right] \\
&= \frac{1}{2i}\left[\boldsymbol{\Sigma}, \mathcal{H}^{\rm EM}_{\rm D}\right] + \frac{1}{2E_p{}'}\left[\beta\left(\boldsymbol{p}-e\boldsymbol{A}\right)\times\boldsymbol{\alpha}, \mathcal{H}^{\rm EM}_{\rm D}\right] \\
&\quad - \frac{1}{i}\left[\frac{(\boldsymbol{p}-e\boldsymbol{A})\times(\boldsymbol{\Sigma}\times(\boldsymbol{p}-e\boldsymbol{A}))}{2E_p{}'(E_p{}'+m_0c^2)}, \mathcal{H}^{\rm EM}_{\rm D}\right] \\
&= -c\boldsymbol{\alpha}\times(\boldsymbol{p}-e\boldsymbol{A}) + \frac{c\beta}{E_p{}'}\boldsymbol{p}\times(\boldsymbol{p}-2e\boldsymbol{A}) \\
&\quad + \frac{cp^2}{E_p{}'(E_p{}'+m_0c^2)}\boldsymbol{\alpha}\times(\boldsymbol{p}-e\boldsymbol{A}) + \frac{ce}{4E_p{}'(E_p{}'+m_0c^2)} \\
&\quad \Big[3\boldsymbol{\Sigma}[\boldsymbol{B}\cdot(\boldsymbol{\alpha}\times\boldsymbol{p})] - (\boldsymbol{\alpha}\cdot\boldsymbol{p})(\boldsymbol{\Sigma}\times\boldsymbol{B}) - \frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha})\boldsymbol{B}\times\boldsymbol{p}}{3} \\
&\quad + 2\boldsymbol{\alpha}\times[\boldsymbol{L}\times(\boldsymbol{B}\times\boldsymbol{p})] + (\boldsymbol{B}\cdot\boldsymbol{\Sigma})(\boldsymbol{\alpha}\times\boldsymbol{p}) \\
&\quad + 2(\boldsymbol{p}\times\boldsymbol{\alpha})(\boldsymbol{B}\cdot\boldsymbol{L})\Big] + \mathfrak{O}(\mathrm{e}^2A^2).
\end{aligned} \tag{30}$$

The derivation followed from the three fundamental commutation relations: $\left[\sigma_i,\sigma_j\right]_- = 2i\epsilon_{ijk}\sigma_k$; $\{\sigma_i,\sigma_j\}_+ = 2\delta_{ij}\boldsymbol{I}_{2\times2}$ and $[r_i,p_j] = i\delta_{ij}$, with $I_{2\times2}$ the 2×2 identity matrix. We mention that the higher-order terms $\mathfrak{O}(\mathrm{e}^2A^2)$ denote that those terms have $\boldsymbol{A}^2$ with a pre-factor of either $E_p{}'$ or $E_p{}'(E_p{}'+m_0c^2)$ in the front. As we describe the spin dynamics in an external field, the definition of E'_p takes the form $E'_p = \sqrt{(\boldsymbol{p}-e\boldsymbol{A})^2c^2+m_0^2c^4}$. The above-derived dynamics has been obtained with the FW spin operator in the Dirac representation. However, it can be also seen as the spin dynamics that results from the inverse relativistic FW transformation of the spin operator $\boldsymbol{S}_{\rm FW}$ with the following FW operator [29, 49, 50]

$$U^{-1}_{\rm FW} = \frac{E'_p+m_0c^2-\beta\mathcal{O}}{\sqrt{2E'_p(E'_p+m_0c^2)}} = \frac{E'_p+m_0c^2-c\beta\boldsymbol{\alpha}\cdot(\boldsymbol{p}-e\boldsymbol{A})}{\sqrt{2E'_p(E'_p+m_0c^2)}}. \tag{31}$$

After the first FW transformation of $\mathcal{H}^{\rm EM}_{\rm D}$ (equation (25)), the Hamiltonian becomes $\mathcal{H}' = \beta E'_p + \mathcal{E}' + \mathcal{O}'$, where one finds the following expressions [29, 50]

$$\mathcal{E}' = i\frac{\partial}{\partial t} + \mathcal{X}\mathcal{F}\mathcal{X} - \mathcal{Y}\mathcal{F}\mathcal{Y}, \tag{32}$$

$$\mathcal{O}' = \mathcal{Y}\mathcal{F}\mathcal{X} - \mathcal{X}\mathcal{F}\mathcal{Y}, \tag{33}$$

where we define $\mathcal{X} = \frac{E'_p+m_0c^2}{\sqrt{2E'_p\left(E'_p+m_0c^2\right)}}$, $\mathcal{Y} = \frac{\beta\mathcal{O}}{\sqrt{2E'_p\left(E'_p+m_0c^2\right)}}$ and $\mathcal{F}$ has already been defined earlier as $\mathcal{F} = \mathcal{E} - i\frac{\partial}{\partial t}$. Note that the odd terms are small compared to the shifted energy E'_p and the original Dirac Hamiltonian in an external field. We have restricted the spin dynamics up to the first-order in field. In the weak field approximation, the higher-order field terms do not contribute significantly. Moreover, as seen in $\mathcal{X}$ and $\mathcal{Y}$, the denominator of such higher-order terms are $E'_p\left(E'_p+m_0c^2\right)$. It is evident that when $\boldsymbol{A}=0$, $\boldsymbol{B}=0$ in equation (30), the spin dynamics in equation (8) is recovered. The meaning of the dynamical terms are explained in the following way. The first term already explains the coupling dynamics for particles and antiparticles. The second term determines the individual dynamics without coupling, however, if $\boldsymbol{A} = 0$, this dynamics vanishes because the curl of a gradient is always zero. More importantly, this term does not involve spins because of the fact that the Dirac matrices $\boldsymbol{\alpha}$ and β anti-commute with each other. The rest of the dynamical terms in equation (30) are due to the relativistic part of the FW spin operator i.e., the last term of equation (6). We note that these terms involve, not only, the spins, but also, the product of spins in the dynamics. One of such terms constitutes as $\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha}$, which can be recast as $\sigma_i^2 = 3\,I_{2\times2}$ (assuming Einstein summation convention). Therefore, this dynamical term does actually not depend on the spins. Similarly, the other terms containing products of spins can be recast as $\sigma_i\sigma_j = \delta_{ij}I_{2\times2} + i\epsilon_{ijk}\sigma_k$, where the first part is again spin independent, while the second part explicitly depends on spins. We conclude for the FW spin-operator dynamics that, along with the spin-dependent dynamics, there are also the spin-independent parts that contribute to the relativistic spin-operator dynamics.

4.1.2. FW transformation of the Dirac Hamiltonian with EM field. Next, we evaluate the FW spin-operator dynamics with the FW transformed Hamiltonian in equation (29). The calculated spin dynamics is

$$\begin{aligned}\frac{\mathrm{d}\boldsymbol{S}_{\mathrm{FW}}}{\mathrm{d}t} &= \frac{1}{i}\left[\boldsymbol{S}_{\mathrm{FW}}, \mathcal{H}^{\mathrm{spin}}_{\mathrm{direct}}\right]\\ &= \frac{1}{2i}\left[\boldsymbol{\Sigma}, \mathcal{H}^{\mathrm{spin}}_{\mathrm{direct}}\right] + \frac{1}{2E_p{}'}\left[\beta\,(\boldsymbol{p}-e\boldsymbol{A})\times\boldsymbol{\alpha}, \mathcal{H}^{\mathrm{spin}}_{\mathrm{direct}}\right]\\ &\quad - \frac{1}{i}\left[\frac{(\boldsymbol{p}-e\boldsymbol{A})\times(\boldsymbol{\Sigma}\times(\boldsymbol{p}-e\boldsymbol{A}))}{2E_p{}'(E_p{}'+m_0c^2)}, \mathcal{H}^{\mathrm{spin}}_{\mathrm{direct}}\right]\\ &= \frac{e\beta}{2m_0}\boldsymbol{\Sigma}\times\boldsymbol{B} + \frac{1}{E_p{}'}\left[\frac{\boldsymbol{p}\times\boldsymbol{\alpha}\left(p^2 - e\boldsymbol{B}\cdot\boldsymbol{L}\right)}{2m_0}\right.\\ &\quad \left. + \frac{ie(\boldsymbol{B}\times\boldsymbol{p})\times\boldsymbol{\alpha}}{4m_0} + \frac{e(\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha})\boldsymbol{B}\times\boldsymbol{p}}{6m_0}\right]\\ &\quad + \frac{e\beta}{2E_p{}'(E_p{}'+m_0c^2)}\left[\frac{[\boldsymbol{B}\cdot(\boldsymbol{p}\times\boldsymbol{\Sigma})]\boldsymbol{p}}{2m_0}\right.\\ &\quad \left. - \frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{p})(\boldsymbol{B}\times\boldsymbol{p})}{2m_0} - \frac{\boldsymbol{p}\times[(\boldsymbol{\Sigma}\times\boldsymbol{B})\times\boldsymbol{p}]}{2m_0}\right]\\ &\quad + \frac{e}{4m_0^2c^2}\left[\boldsymbol{\Sigma}\times(\boldsymbol{E}\times\boldsymbol{p}) + i\frac{(\boldsymbol{p}\times\boldsymbol{\alpha})\times(\boldsymbol{E}\times\boldsymbol{p})}{E_p{}'}\right.\\ &\quad \left. - \frac{\boldsymbol{p}\times[(\boldsymbol{\Sigma}\times[\boldsymbol{E}\times\boldsymbol{p}])\times\boldsymbol{p}]}{E_p{}'(E_p{}'+m_0c^2)}\right] - \frac{ie}{8m_0^2c^2}\\ &\quad \times\left[\boldsymbol{\Sigma}\times\dot{\boldsymbol{B}} + i\frac{(\boldsymbol{p}\times\boldsymbol{\alpha})\times\dot{\boldsymbol{B}}}{E_p{}'} - \frac{\boldsymbol{\alpha}\times\left[\dot{\boldsymbol{B}}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})\right]}{2E_p{}'}\right.\\ &\quad \left. + \frac{\boldsymbol{p}\times\left[(\boldsymbol{\Sigma}\times\dot{\boldsymbol{B}})\times\boldsymbol{p}\right]}{E_p{}'(E_p{}'+m_0c^2)}\right] - \frac{e}{16m_0^3c^4}\\ &\quad \times\left[\beta\boldsymbol{\Sigma}\times\ddot{\boldsymbol{B}} + \frac{(\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha})\ddot{\boldsymbol{B}}\times\boldsymbol{p}}{3E_p{}'} + \beta\frac{\boldsymbol{p}\times\left[(\boldsymbol{\Sigma}\times\ddot{\boldsymbol{B}})\times\boldsymbol{p}\right]}{E_p{}'(E_p{}'+m_0c^2)}\right]\\ &\quad + \mathfrak{O}(\mathrm{e}^2A^2). \qquad (34)\end{aligned}$$

As we have started from a semi-relativistic expansion of the Dirac Hamiltonian, it is evidently diagonal in the particle–antiparticle Hilbert space. However, the calculated spin dynamics suggests that the particle–antiparticle coupling terms (off-diagonal) are nonetheless important, when one considers the relativistic FW spin operator. Furthermore, the spin dynamics shows the importance of spin-independent terms in the Hamiltonian in equation (29). The kinetic energy term in equation (29) is explicitly spin independent, however, this term contributes to the spin dynamics due to the form of the relativistic spin operator. In fact, the commutator $[\beta\boldsymbol{p}\times\boldsymbol{\alpha}, \beta\boldsymbol{p}\cdot\boldsymbol{p}]$ leads to an anti-commutator $\{\boldsymbol{p}\times\boldsymbol{\alpha}, \boldsymbol{p}\cdot\boldsymbol{p}\}$ because the Dirac matrices $\boldsymbol{\alpha}$ and β anti-commute with each other and contribute to the dynamical equation of motion. The diagonal terms in equation (34) have useful meanings as discussed in the context of magnetization dynamics [24, 26, 34]. The first term $\boldsymbol{\Sigma}\times\boldsymbol{B}$ signifies the precession of a single spin around a field, the terms $\boldsymbol{\Sigma}\times(\boldsymbol{E}\times\boldsymbol{p})$ and $\boldsymbol{\Sigma}\times\dot{\boldsymbol{B}}$ explains the energy dissipation in terms of damping processes [7, 8]. The higher-order energy dissipation terms stem from the relativistic parts of the spin operator. These terms can be identified as the last terms in the second and third lines of equation (34). The other terms in the second and third lines of equation (34) are evidently off-diagonal, thus, they pertain to the particle-antiparticle interactions. Higher order relativistic spin dynamical terms can be noticed from the last line of equation (34). Such terms have been associated with spin dynamics in the inertial regime [51–54], which is a higher-order relativistic spin–orbit coupling effect [34, 35]. Note that the dynamical term with $\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha}$ can be seen as a spin-independent term as described previously. Overall, the spin dynamics with the relativistic FW spin operator exhibits a dynamics that has two contributions: (1) spin-dependent and (2) spin-independent terms.

4.2. Pryce spin operator

Next, we consider the Pryce spin operator [12, 13]. In the presence of an electromagnetic field, the Pryce spin operator, equation (15), can be written simply by replacing $\boldsymbol{p}\to(\boldsymbol{p}-e\boldsymbol{A})$. Thus, the Pryce spin operator becomes field dependent [4].

4.2.1. Dirac Hamiltonian with an EM field. The spin dynamics with the Dirac Hamiltonian in the presence of an EM field is rather different and calculated as

$$\begin{aligned}\frac{\mathrm{d}\boldsymbol{S}_{\mathrm{Py}}}{\mathrm{d}t} &= \frac{1}{i}\left[\boldsymbol{S}_{\mathrm{Py}}, \mathcal{H}^{\mathrm{EM}}_{\mathrm{D}}\right]\\ &= \frac{1}{2i}\left[\beta\boldsymbol{\Sigma}, \mathcal{H}^{\mathrm{EM}}_{\mathrm{D}}\right]\\ &\quad + \frac{1}{2i}\left[(1-\beta)\frac{[\boldsymbol{\Sigma}\cdot(\boldsymbol{p}-e\boldsymbol{A})](\boldsymbol{p}-e\boldsymbol{A})}{(p-eA)^2}, \mathcal{H}^{\mathrm{EM}}_{\mathrm{D}}\right]\\ &= \frac{ce}{2(p-eA)^2}(\boldsymbol{\alpha}\cdot\boldsymbol{p})(\boldsymbol{\Sigma}\times\boldsymbol{B})\\ &\quad + \frac{ce\beta}{3i(p-eA)^2}(\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha})(\boldsymbol{B}\cdot\boldsymbol{L})\boldsymbol{p} + \mathfrak{O}(\mathrm{e}^2A^2). \qquad (35)\end{aligned}$$

In the above derivation, the first commutator $[\beta\mathbf{\Sigma}, c\boldsymbol{\alpha}\cdot\boldsymbol{p}]$ exactly cancels the last commutator $[\beta\frac{(\mathbf{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}, c\boldsymbol{\alpha}\cdot\boldsymbol{p}]$. However, the field-dependent terms of those commutators contribute to the second term of equation (35) in the linear order. The linear order field-dependent terms of $[\frac{(\mathbf{\Sigma}\cdot\boldsymbol{p})\boldsymbol{p}}{p^2}, c\boldsymbol{\alpha}\cdot\boldsymbol{p}]$ result in the first term of equation (35). Note that in the absence of the EM field i.e., $\boldsymbol{B}=0$, the dynamics in equation (35) recovers the spin dynamics for a free Dirac particle in equation (17). It is interesting to point out that the dynamics in equation (35) contains only the off-diagonal elements in the matrix formalism. The latter means that this dynamics is governed by the coupling between the particles and antiparticles which comes from the Dirac Hamiltonian itself, the term $\boldsymbol{\alpha}\cdot\boldsymbol{p}$. This feature of the Pryce spin dynamics stands in contrast to the FW spin dynamics in equation (30), where both particle diagonal and off-diagonal terms contribute. In fact, the FW spin dynamics contains some terms with only diagonal contributions. The first term in equation (35) is notably off-diagonal and can be represented by $\sigma_i\sigma_j$. Following the similar argument, this term can be split into a spin-independent part and a spin-dependent part. Thus, the Pryce spin dynamics contains also spin dependent and independent contributions, like the FW spin dynamics as discussed earlier.

4.2.2. FW transformation of the Dirac Hamiltonian with an EM field. Next, we calculate the spin dynamics from the transformed Hamiltonian in equation (29). The derived dynamical equation is

$$
\begin{aligned}
\frac{\mathrm{d}\boldsymbol{S}_{\mathrm{Py}}}{\mathrm{d}t} &= \frac{1}{i}\left[\boldsymbol{S}_{\mathrm{Py}}, \mathcal{H}^{\mathrm{spin}}_{\mathrm{direct}}\right] \\
&= \frac{1}{2i}\left[\beta\mathbf{\Sigma}, \mathcal{H}^{\mathrm{spin}}_{\mathrm{direct}}\right] \\
&\quad + \frac{1}{2i}\left[(1-\beta)\frac{[\mathbf{\Sigma}\cdot(\boldsymbol{p}-e\boldsymbol{A})](\boldsymbol{p}-e\boldsymbol{A})}{(p-eA)^2}, \mathcal{H}^{\mathrm{spin}}_{\mathrm{direct}}\right] \\
&= \frac{e}{2m_0}\mathbf{\Sigma}\times\boldsymbol{B} + \frac{e(1-\beta)}{2m_0(p-eA)^2}(\mathbf{\Sigma}\cdot\boldsymbol{p})(\boldsymbol{B}\times\boldsymbol{p}) \\
&\quad + \frac{e\beta}{4m_0^2c^2}\mathbf{\Sigma}\times(\boldsymbol{E}\times\boldsymbol{p}) + \frac{e(1-\beta)}{8m_0^2c^2(p-eA)^2} \\
&\quad \times\Big[(\mathbf{\Sigma}\cdot\boldsymbol{p})(\mathbf{\Sigma}\cdot\dot{\boldsymbol{B}})\boldsymbol{p} - (\mathbf{\Sigma}\cdot\boldsymbol{p})(\boldsymbol{L}\cdot\dot{\boldsymbol{B}})\boldsymbol{p} \\
&\quad - \frac{\Sigma^2\boldsymbol{p} + (\mathbf{\Sigma}\cdot\boldsymbol{p})\mathbf{\Sigma}}{2}(\dot{\boldsymbol{B}}\cdot\boldsymbol{p})\Big] - \frac{ie\beta}{8m_0^2c^2} \\
&\quad \times\left[\mathbf{\Sigma}\times\dot{\boldsymbol{B}} + \beta(1-\beta)\frac{[(\mathbf{\Sigma}\times\dot{\boldsymbol{B}})\cdot\boldsymbol{p}]\boldsymbol{p}}{(p-eA)^2}\right] - \frac{e}{16m_0^3c^4} \\
&\quad \times\left[\mathbf{\Sigma}\times\ddot{\boldsymbol{B}} + \beta(1-\beta)\frac{[(\mathbf{\Sigma}\times\ddot{\boldsymbol{B}})\cdot\boldsymbol{p}]\boldsymbol{p}}{(p-eA)^2}\right] + \mathfrak{O}(e^2A^2).
\end{aligned}
\tag{36}
$$

As we have started from a diagonalized Hamiltonian and the Pryce spin operator which is diagonal, too, all the derived dynamical terms are diagonal as well. This means that the corresponding dynamics only describes the particles and antiparticles, not the coupling between them. To derive such dynamics, one has to note that the kinetic energy does commute with the first term of the Pryce spin operator in equation (15), however, it does not commute with the second term because the latter contains the momentum operator as well. Such non-commutator implies that *not only* the spin, *but also* the orbital momentum contributes to the relativistic spin dynamics through the spin–orbit coupling-like mechanisms that is considered in the relativistic spin operator of Pryce type. The dynamical terms in equation (36) can be related to similar terms as derived in equation (34). For example, the first term in equation (36) describes the spin precession around a field, the third term and the first terms of third line in equation (36) explain the energy dissipation from spin to other degrees of freedom. The first term of the last line in equation (36) accounts for the spin dynamics in the inertial regime. The other remaining terms in equation (36) do not directly correspond to the FW dynamics in equation (34). However, they derive from the relativistic part of the Pryce spin operator. Thus, they contain either $(1-\beta)$ or $\beta(1-\beta)$ as appear in equation (36).

5. Summary and discussions

Traditionally, the spin dynamics is derived for the nonrelativistic spin operator (see, e.g., [24]). Here, we have derived the spin dynamics with relativistic spin operators. We have used three different Hamiltonians to derive the corresponding spin dynamics: (1) free-particle Dirac Hamiltonian, (2) Dirac Hamiltonian in an EM environment, and (3) diagonalized Dirac Hamiltonian in the presence of an EM field. The relativistic spin dynamics is a constant of motion when the free-particle Dirac Hamiltonian is considered. This result however only holds for relativistic spin operators of FW and Pryce type, making them ideal candidates for proper relativistic spin operators. These two relativistic spin operators are however very different: the FW spin operator has diagonal and off-diagonal elements in particle-antiparticle space, whereas, the Pryce operator has only diagonal elements. These spin operators involve not only spin angular momentum in terms of Pauli spin matrices, but also, the orbital angular momentum in terms of momentum operator $\boldsymbol{p}$. The derived dynamics of these operators in an EM field provides two important informations: (1) the particle–antiparticle coupling terms contribute to the spin dynamics, even if one starts with a diagonalized Hamiltonian, (2) there exist two separate parts (spin-dependent and spin-independent terms) of the derived spin dynamics. We note that some dynamical terms appear in both the FW and Pryce spin dynamics in similar way, however, due of the relativistic spin operators' construction, additional terms exist. The derived dynamics reveals that coupling of the orbital angular momentum with spin contributes to the spin dynamics, moreover, a few dynamical terms *only* depend on the orbital angular momentum. To compare the relativistic spin dynamics, we provide tables for the two calculated spin dynamics in the appendix A. The FW and Pryce spin dynamical terms have been compared, for the Dirac Hamiltonian with an EM field in table A1 and for FW transformation of the Dirac Hamiltonian with an EM field in table A2. Note that we have not included all the terms of equations (36) and (34) in the tables. The reason is that some terms can be recast to show that they are equivalent.

Several terms in the derived spin dynamics equations of the two considered proper spin operators are rather distinct. The FW spin operator has diagonal and off-diagonal components which means it accounts for the coupling terms in the particle-antiparticle Hilbert space. When we compare the two equations for spin motion, equation (34) for FW dynamics and equation (36) for Pryce dynamics, which have been derived from the same Hamiltonian, we observe that the Pryce dynamics in equation (36) is diagonal and does not involve such coupling terms. In fact, the Pryce dynamics involves terms which have $(1-\beta)$ that translates to zero contribution for the upper component in the 2×2 formalism. Therefore, the 2×2 Pryce dynamics recovers exactly the same dynamical terms as the Pauli spin dynamics [34]. As already mentioned, the FW dynamics in equation (34) contains particle diagonal as well as off-diagonal terms. To achieve a 2×2 electron spin dynamics, one has to diagonalize. Even then, the additional terms appear apart from the standard Pauli spin dynamics. Moreover, the additional terms account for spin angular momentum and orbital contributions as well. Therefore, one can conclude that for the spin dynamics in an applied EM field, the two spin operators have their own validity regime. We thus consider three operational field regimes: weak, intermediate, and strong. In the weak field regime, the Pauli spin operator can describe the spin dynamics, while, for an intermediate field regime where the spin–orbit coupling is important, the Pryce spin operator seems to describe the proper spin dynamics. However, in the stronger field regime, where the spin–orbit and relativistic particle-antiparticle couplings are present, the FW spin operator suits the best for describing the spin dynamics. The derived operational spin dynamics regimes of the Pauli, Pryce and FW spin operators are schematically summarized as follows: considering *relativity*, which in relativistic theory, is given through the particle velocity (v) being closer to the velocity of light c. At the ultra-relativistic limit $v\to c$, the particle-antiparticle coupling exists, which can be well described by the FW spin dynamics. On the other hand, at the limit $v<c$, the particle–antiparticle coupling can be neglected and therefore the corresponding spin dynamics can be captured by the Pryce spin operator. Finally, at the nonrelativistic limit $v\ll c$, the Pauli spin operator describes the spin operator well enough.

Finally, one can observe that the derived equations of motion can be experimentally investigated as described in the following. In extended Landau–Lifshitz–Gilbert (LLG) spin dynamics, the Gilbert damping and inertial dynamical equation of motion of a spin (without the spin precession) are commonly described as

$$\frac{\mathrm{d}\boldsymbol{S}}{\mathrm{d}t} = \underbrace{\alpha\boldsymbol{S}\times\frac{\mathrm{d}\boldsymbol{S}}{\mathrm{d}t}}_{\text{Gilbert damping}} + \underbrace{\iota\boldsymbol{S}\times\frac{\mathrm{d}^2\boldsymbol{S}}{\mathrm{d}t^2}}_{\text{Inertial dynamics}}\,, \tag{37}$$

where $\boldsymbol{S}$ defines the spin vector representing a three-dimensional and time-dependent Pauli spin operator and α and ι denote the Gilbert damping and magnetic inertial parameters, respectively. These contributions stem from the dynamical terms $\boldsymbol{S}\times\dot{\boldsymbol{B}}$ and $\boldsymbol{S}\times\ddot{\boldsymbol{B}}$ that have previously been obtained while starting from the nonrelativistic Pauli spin operator and working with the extended Pauli Hamiltonian [8, 34]. We note that we have already obtained equivalent terms in our derived equation of motion, e.g., equations (34) and (36) in the form of $\boldsymbol{\Sigma}\times\dot{\boldsymbol{B}}$ and $\boldsymbol{\Sigma}\times\ddot{\boldsymbol{B}}$, respectively. In this article, the consideration of relativistic spin operators shows that there can be *additional* contributions to these dynamical terms. For example, when we consider the last terms of the last two lines in equation (34), one can show that these provide the following dynamics

$$\frac{\mathrm{d}\boldsymbol{S}_{\mathrm{FW}}}{\mathrm{d}t} \propto \frac{p^2}{E_p'(E_p'+m_0c^2)}\left[(\boldsymbol{\Sigma}\times\dot{\boldsymbol{B}})+\beta(\boldsymbol{\Sigma}\times\ddot{\boldsymbol{B}})\right] - \frac{\boldsymbol{p}}{E_p'(E_p'+m_0c^2)}\left[\left[\boldsymbol{p}\cdot(\boldsymbol{\Sigma}\times\dot{\boldsymbol{B}})\right]+\beta\left[\boldsymbol{p}\cdot(\boldsymbol{\Sigma}\times\ddot{\boldsymbol{B}})\right]\right]. \tag{38}$$

It is clear that the first and third right-hand terms contribute to the Gilbert damping dynamics, however, the second and fourth terms contribute to the magnetic inertial dynamics. Note that these terms can be seen as higher-order contributions to the spin dynamics. As an outlook, it would be inspiring to experimentally verify the existence of such relativistic corrections to the damping and inertial spin dynamics.

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Appendix A. Comparison of spin dynamical terms

In this Appendix, we provide detailed descriptions of several of the appearing spin dynamical terms in the equations for spin motion derived in the article. The comparison tables include the dynamical terms, their origin in the calculated commutators, whether they are present in the FW and/or Pryce dynamics and comments on the dynamical terms. In the table A1, we compare the two dynamical equations i.e., equations (30) and (35) for the fully relativistic Hamiltonian in the presence of an external field. We note that the FW spin dynamical equation in equation (30) contains similar terms as the Pryce spin dynamics in equation (35). However, there are additional dynamical terms present in the FW dynamics due to the fact that the FW spin operator includes the influence of particle-antiparticle coupling.

In table A2, we compare the two spin dynamical equations, i.e., equations (34) and (36) for the relativistic direct

Table A1. Comparison of several occurring dynamical terms in the FW spin dynamics (equation (30)) and the Pryce spin dynamics (equation (35)). Diagonal and off-diagonal refer to the particle-antiparticle Hilbert space.

Dirac Hamiltonian with an EM field				
Spin dynamical terms	Origin	In FW dynamics (equation (30))	In Pryce dynamics (equation (35))	Comments
$\boldsymbol{\alpha}\times\boldsymbol{A}$	$[\boldsymbol{\Sigma}, \boldsymbol{\alpha}\cdot\boldsymbol{A}]$	✓	✗	Off-diagonal, spin dependent
$\beta\boldsymbol{p}\times\boldsymbol{A}$	$[\beta\boldsymbol{p}\times\boldsymbol{\alpha}, \boldsymbol{\alpha}\cdot\boldsymbol{A}]$	✓	✗	Diagonal, spin independent
$(\boldsymbol{p}\times\boldsymbol{\alpha})(\boldsymbol{B}\cdot\boldsymbol{L})$	$[\boldsymbol{A}\times(\boldsymbol{\Sigma}\times\boldsymbol{p}), \boldsymbol{\alpha}\cdot\boldsymbol{p}]$	✓	✗	Off-diagonal, spin dependent
$(\boldsymbol{\alpha}\cdot\boldsymbol{p})(\boldsymbol{\Sigma}\times\boldsymbol{B})$	$[(\boldsymbol{\Sigma}\cdot\boldsymbol{A})\boldsymbol{p}, \boldsymbol{\alpha}\cdot\boldsymbol{p}]$ and similar terms (Pryce) $[\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{A}), \boldsymbol{\alpha}\cdot\boldsymbol{p}]$ and similar terms (FW)	✓	✓	Off-diagonal, spin independent and spin dependent
$(\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha})\boldsymbol{B}\times\boldsymbol{p}$	$[\beta(\boldsymbol{\Sigma}\cdot\boldsymbol{A})\boldsymbol{p}, \boldsymbol{\alpha}\cdot\boldsymbol{p}]$ and similar terms (Pryce) $[\boldsymbol{p}\times(\boldsymbol{\Sigma}\times\boldsymbol{A}), \boldsymbol{\alpha}\cdot\boldsymbol{p}]$ and similar terms (FW)	✓	✓	Off-diagonal, spin independent Off-diagonal, spin independent

Table A2. Comparison of several of the occurring dynamical terms in the FW (equation (34)) and Pryce (equation (36)) spin dynamics.

FW transformed Dirac Hamiltonian with an EM field				
Spin dynamical terms	Origin	In FW dynamics (equation (34))	In Pryce dynamics (equation (36))	Comments
$(\boldsymbol{p}\times\boldsymbol{\alpha})(\boldsymbol{B}\cdot\boldsymbol{L})$	$[\beta\boldsymbol{p}\times\boldsymbol{\alpha}, \boldsymbol{A}\cdot\boldsymbol{p}]$	✓	✗	Off-diagonal, spin dependent
$(\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha})\boldsymbol{B}\times\boldsymbol{p}$	$[\beta\boldsymbol{p}\times\boldsymbol{\alpha}, \beta\boldsymbol{\Sigma}\cdot\boldsymbol{B}]$	✓	✗	Off-diagonal, spin independent
$(\boldsymbol{p}\times\boldsymbol{\alpha})\times\dot{\boldsymbol{B}}$	$[\beta\boldsymbol{p}\times\boldsymbol{\alpha}, \boldsymbol{\Sigma}\cdot(\boldsymbol{E}\times\boldsymbol{p})]$	✓	✗	Off-diagonal, spin dependent
$\boldsymbol{\alpha}\times[\dot{\boldsymbol{B}}\times(\boldsymbol{\Sigma}\times\boldsymbol{p})]$	$[\beta\boldsymbol{p}\times\boldsymbol{\alpha}, \boldsymbol{\Sigma}\cdot(\boldsymbol{E}\times\boldsymbol{p})]$	✓	✗	Off-diagonal, spin independent and spin dependent
$(\boldsymbol{\Sigma}\cdot\boldsymbol{\alpha})\ddot{B}\times\boldsymbol{p}$	$[\beta\boldsymbol{p}\times\boldsymbol{\alpha}, \beta\boldsymbol{\Sigma}\cdot\ddot{\boldsymbol{B}}]$	✓	✗	Off-diagonal, spin independent
$(\boldsymbol{p}\times\boldsymbol{\alpha})(\boldsymbol{E}\times\boldsymbol{p})$	$[\beta\boldsymbol{p}\times\boldsymbol{\alpha}, \boldsymbol{\Sigma}\cdot(\boldsymbol{E}\times\boldsymbol{p})]$	✓	✗	Off-diagonal, spin dependent
$(\boldsymbol{\Sigma}\cdot\boldsymbol{p})(\boldsymbol{B}\times\boldsymbol{p})$	$[\boldsymbol{A}\times(\boldsymbol{\Sigma}\times\boldsymbol{p}), p^2]$ and similar terms (FW) $[(\boldsymbol{\Sigma}\cdot\boldsymbol{A})\boldsymbol{p}, p^2]$ and similar terms (Pryce)	✓	✓	Diagonal, spin dependent Diagonal, spin dependent
$\boldsymbol{\Sigma}\times(\boldsymbol{E}\times\boldsymbol{p})$	$[\boldsymbol{\Sigma}, \boldsymbol{\Sigma}\cdot(\boldsymbol{E}\times\boldsymbol{p})]$	✓	✓	Diagonal, spin dependent
$\boldsymbol{\Sigma}\times\boldsymbol{B}$	$[\boldsymbol{\Sigma}, \boldsymbol{\Sigma}\cdot\boldsymbol{B}]$	✓	✓	Diagonal, spin dependent
$\boldsymbol{\Sigma}\times\dot{\boldsymbol{B}}$	$[\boldsymbol{\Sigma}, \boldsymbol{\Sigma}\cdot\dot{\boldsymbol{B}}]$	✓	✓	Diagonal, spin dependent
$\boldsymbol{\Sigma}\times\ddot{\boldsymbol{B}}$	$[\boldsymbol{\Sigma}, \boldsymbol{\Sigma}\cdot\ddot{\boldsymbol{B}}]$	✓	✓	Diagonal, spin dependent

spin-field coupling Hamiltonian in the presence of an external field. Again, the FW spin dynamics contains several dynamical terms that are not present in the Pryce dynamics.

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References

[1] Newton T D and Wigner E P 1949 *Rev. Mod. Phys.* **21** 400–6
[2] Jordan T F and Mukunda N 1963 *Phys. Rev.* **132** 1842–8
[3] Lorente M and Roman P 1974 *J. Math. Phys.* **15** 70–4
[4] Bauke H, Ahrens S, Keitel C H and Grobe R 2014 *Phys. Rev.* A **89** 052101
[5] Bliokh K Y, Dennis M R and Nori F 2017 *Phys. Rev.* A **96** 023622
[6] O'Connell R F and Wigner E P 1997 *Position Operators for Systems Exhibiting the Special Relativistic Relation between Momentum and Velocity* (Berlin: Springer) pp 335–7
[7] Hickey M C and Moodera J S 2009 *Phys. Rev. Lett.* **102** 137601
[8] Mondal R, Berritta M and Oppeneer P M 2016 *Phys. Rev.* B **94** 144419
[9] Atxitia U, Hinzke D and Nowak U 2016 *J. Phys. D: Appl. Phys.* **50** 033003
[10] Foldy L L and Wouthuysen S A 1950 *Phys. Rev.* **78** 29–36
[11] Foldy L L 1952 *Phys. Rev.* **87** 688–93
[12] Pryce M H L 1935 *Proc. R. Soc.* A **150** 166–72
[13] Pryce M H L 1948 *Proc. R. Soc.* A **195** 62–81
[14] Dirac P A M 1928 *Proc. R. Soc.* A **117** 610–24
[15] Dirac P A M 1928 *Proc. R. Soc.* A **118** 351–61
[16] Dirac P A M 1930 *Proc. R. Soc.* A **126** 360
[17] Strange P 1998 *Relativistic Quantum Mechanics: With Applications in Condensed Matter and Atomic Physics* (Cambridge: Cambridge University Press)
[18] Bauke H, Ahrens S, Keitel C H and Grobe R 2014 *New J. Phys.* **16** 043012
[19] Caban P, Rembieliński J and Włodarczyk M 2013 *Phys. Rev.* A **88** 022119
[20] Deriglazov A A and Ramírez W G 2017 *Adv. Math. Phys.* **2017** 7397159
[21] Czachor M 1997 *Phys. Rev.* A **55** 72–7
[22] de Vries E and Jonker J E 1968 *Nucl. Phys.* B **6** 213–25
[23] Greiner W 2000 *Relativistic Quantum Mechanics. Wave Equations* (Berlin: Springer)

[24] Mondal R, Berritta M and Oppeneer P M 2018 *Phys. Rev.* B **98** 214429
[25] Bjorken J and Drell S 1964 *Relativistic Quantum Mechanics* (New York: McGraw-Hill)
[26] Mondal R 2017 Relativisitic theory of laser-induced magnetization dynamics *PhD Thesis* Uppsala University, Uppsala
[27] de Vries E 1970 *Fortschr. Phys.* **18** 149–82
[28] Silenko A J 2016 *Phys. Rev.* A **94** 032104
[29] Silenko A J 2003 *J. Math. Phys.* **44** 2952–66
[30] Silenko A J 2016 *Phys. Rev.* A **93** 022108
[31] Silenko A J 2015 *Phys. Rev.* A **91** 022103
[32] Mondal R, Berritta M, Carva K and Oppeneer P M 2015 *Phys. Rev.* B **91** 174415
[33] Mondal R, Berritta M, Paillard C, Singh S, Dkhil B, Oppeneer P M and Bellaiche L 2015 *Phys. Rev.* B **92** 100402(R)
[34] Mondal R, Berritta M, Nandy A K and Oppeneer P M 2017 *Phys. Rev.* B **96** 024425
[35] Mondal R, Berritta M and Oppeneer P M 2018 *J. Phys.: Condens. Matter* **30** 265801
[36] Hinschberger Y and Hervieux P A 2012 *Phys. Lett.* A **376** 813–9
[37] Zawadzki W 2005 *Am. J. Phys.* **73** 756–8
[38] Kraft T, Oppeneer P M, Antonov V N and Eschrig H 1995 *Phys. Rev.* B **52** 3561–70
[39] Crépieux A and Bruno P 2001 *Phys. Rev.* B **64** 094434
[40] Mondal R, Berritta M and Oppeneer P M 2017 *J. Phys.: Condens. Matter* **29** 194002
[41] Paillard C, Mondal R, Berritta M, Dkhil B, Singh S, Oppeneer P M and Bellaiche L 2016 *Proc. SPIE* **9931** 99312E
[42] Mondal R, Berritta M, Nandy A K and Oppeneer P M 2018 *Proc. SPIE* **10732** 107322E
[43] Mondal R, Donges A, Ritzmann U, Oppeneer P M and Nowak U 2019 *Phys. Rev.* B **100** 060409
[44] Bauke H, Ahrens S, Keitel C H and Grobe R 2014 *New J. Phys.* **16** 103028
[45] Bauke H, Ahrens S and Grobe R 2014 *Phys. Rev.* A **90** 052101
[46] Aleksandrov I A, Tumakov D A, Kudlis A, Shabaev V M and Rosanov N N 2020 Relativistic electron spin dynamics in a strong unipolar laser field (arXiv:2005.02839)
[47] de Groot S R and Suttorp L G 1968 *Physica* **39** 77–83
[48] de Groot S R and Suttorp L G 1972 *Foundations of Electrodynamics* (Amsterdam: North-Holland)
[49] Suttorp L G and De Groot S R 1970 *Il Nuovo Cimento* A **65** 245–74
[50] Silenko A Y 2013 *Theor. Math. Phys.* **176** 987–99
[51] Wegrowe J E and Ciornei M C 2012 *Am. J. Phys.* **80** 607–11
[52] Ciornei M C, Rubí J M and Wegrowe J E 2011 *Phys. Rev.* B **83** 020410
[53] Wegrowe J E and Olive E 2016 *J. Phys.: Condens. Matter* **28** 106001
[54] Olive E, Lansac Y, Meyer M, Hayoun M and Wegrowe J E 2015 *J. Appl. Phys.* **117** 213904