

Direct mechanism of spin orientation by circularly polarized light

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Dissipative spin dynamics in a rotating magnetic field created by a circularly polarized light is considered. With the generalized Bloch equations we show that the steady-state spin polarization along the laser beam axis arises as a result of the direct transfer of the angular momentum from photons to the spin-1/2 in a medium without optical absorption. An application of this mechanism for explaining the inverse Faraday effect is discussed. [S0163-1829(99)04829-8]

Much attention has been focused in recent years on the study of nonequilibrium spin dynamics in the presence of a dissipative environment.^{1,2} Among other things this interest is caused by the rapid progress of magnetoelectronics³ as well as by the possibility of putting electronic or nuclear spins to work in quantum computers.⁴

The employment of spin-polarized carriers is shared by all magnetoelectronic devices. It is well known⁵ that the simplest way to polarize a spin particle in a given direction is in applying of a constant magnetic field along this direction. A spin orientation by circularly polarized light in semiconductors with optical absorption is also a possibility.^{6,7} In this case the orientation is connected with the interband transitions, and the transfer of the angular momentum from photons to electrons is due to a spin-orbit interaction. The spin-orbit interaction can also account for a spin polarization induced by a circularly polarized light in crystals without a mirror symmetry.⁸

In the present work we analyze a mechanism of the direct transfer of the angular momentum from the circularly polarized light to the spin-1/2 in a medium without optical absorption. To do this we derive non-Markovian equations for the spin particle coupled to the heat bath because the conventional Bloch equations⁵ do not give an insight into the occurrence of spin orientation and a constant magnetization in the field of a circularly polarized laser beam. This optically-induced magnetization is usually supposed to be due to the optical Stark effect which removes the degeneracy of ground states;^{9,10} in so doing to explain this phenomenon known as the inverse Faraday effect¹¹⁻¹³ the interaction of the light with charge degrees of freedom of atoms is taken into consideration. We propose here another mechanism descriptive of the production of a magnetization by circularly polarized light in a nonabsorbing medium. It follows from Maxwell's equations¹⁴ that the circularly polarized laser beam creates not only an electric field, but a rotating magnetic field as well. We consider an influence of this magnetic field on spin degrees of freedom coupled also to a dissipative environment and show that the steady-state spin orientation along the laser beam axis arises in response to the joint action of the rotating magnetic field and the dissipative environment. An investigation of this effect can be of particular importance not only for a better understanding of controlled nonequilibrium spin dynamics in magnetic nanostructures but for laser-enhanced NMR spectroscopy¹⁵ as well.

The Hamiltonian of a quantum particle with a spin-1/2 subjected to the rotating magnetic field $\vec{B}(t) = (B_0 \cos \omega_0 t, \sin \omega_0 t, 0)$ and coupled to a heat bath with variables $\{Q_i(t)\} (i=x, y, z)$ may be written as

$$H = (\Delta/2) (\sigma_x \cos \omega_0 t + \sigma_y \sin \omega_0 t) - \vec{\sigma} \cdot \vec{Q}(t) + H_B. \quad (1)$$

Here H_B is the free Hamiltonian of the heat bath, $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ are the Pauli matrices of the particle with a magnetic moment $\vec{\mu} = g\mu_0\vec{\sigma}$, mass m , and g-factor g ; $\mu_0 = e\hbar/2mc$ is the magneton, and $\Delta = -2g\mu_0 B_0$ is the energy splitting. As mentioned above, the rotating magnetic field $\vec{B}(t)$ can be produced by the circularly polarized laser beam propagating along the z axis. Contrary to the works^{6,7,9-11} devoted to the theory of optical orientation, we ignore here the influence of the laser beam on charge degrees of freedom and suppose that the spatial state of the particle is unaffected. At this stage we do not dwell on the specific model of the heat bath. An important point is that its unperturbed variables $\{Q_i^{(0)}(t)\} (i=x, y, z)$ governed by the free Hamiltonian H_B are described by the Gaussian statistics¹⁶ with a response function

$$\varphi_{ij}(t, t') = \langle i [Q_i^{(0)}(t), Q_j^{(0)}(t')]_- \rangle \theta(t - t'), \quad (2)$$

a covariance

$$M_{ij}(t, t') = \langle \frac{1}{2} [Q_i^{(0)}(t), Q_j^{(0)}(t')]_+ \rangle, \quad (3)$$

and zero mean values $\langle Q_i^{(0)}(t) \rangle = 0$. The Fourier transforms of the response function and covariance, namely, the susceptibility and the spectral density of the heat bath fluctuations, will be designed as $\chi_{ij}(\omega)$ and $S_{ij}(\omega)$, respectively; $\theta(t)$ is the unit Heaviside step function, the brackets $\langle \dots \rangle$ signify an average over the initial state of the heat bath with a temperature T in a combination with the trace over spins ($k_B = 1, \hbar = 1$). As this takes place, the total heat bath variables $Q_i(t)$ are linear in spin operators:¹⁶

$$Q_i(t) = Q_i^{(0)}(t) + \int dt_1 \varphi_{ij}(t, t_1) \sigma_j(t_1). \quad (4)$$

In the frame of reference rotating in synchronism with the external magnetic field the Hamiltonian (1) rearranges to the form

$$H = (\Delta/2)x(t) - x_i(t)P_i(t) + H_B, \quad (5)$$

where the new spin operators are $x_3 = z(t) = \sigma_z$ and $x_i = W_{ij}(t)\sigma_j$, $x_1 = x$, $x_2 = y$, $W_{11} = W_{22} = \cos \omega_0 t$, $W_{12} = -W_{21} = -\sin \omega_0 t$, $P_i = W_{ij}(t)Q_j$ ($i, j = 1, 2$) are the new variables of the heat bath, $P_3 = Q_z$.

The Heisenberg equations for the spin operators in the rotating frame of reference ($i, j, k = 1, 2, 3$) follow from the Hamiltonian (5)

$$\dot{x}_i = \Lambda_{ij}x_j + 2\varepsilon_{ijk}x_jP_k. \quad (6)$$

Here $\Lambda_{12} = -\Lambda_{21} = \omega_0$, $\Lambda_{23} = -\Lambda_{32} = -\Delta$, ε_{ijk} is the anti-symmetric unit tensor, $\varepsilon_{123} = 1$.

In the case of the isotropic dissipative environment we have $\varphi_{ij}(t, t') = \delta_{ij}\varphi(t, t')$, and the heat bath variables P_1, P_2 are linear in spin operators x, y , whereas the heat bath operator P_3 is linear in $z(t)$ with the following response functions in the rotating frame of reference: $\varphi_C(t, t_1) = \varphi(t, t_1)\cos \omega_0(t-t_1)$, $\varphi_S(t, t_1) = \varphi(t, t_1)\sin \omega_0(t-t_1)$.

According to the method developed in Refs. 16 and 17, we can deduce now the stochastic Heisenberg-Langevin equations. But here we are interested in the average dynamics of spin operators only. The corresponding non-Markovian equations can be obtained by substituting of the total heat bath variables into Eq. (6) followed by averaging over the initial state of the heat bath:

$$\begin{aligned} \langle \dot{x}(t) \rangle - \omega_0 \langle y(t) \rangle &= 2 \int dt_1 \bar{M}(t, t_1) \langle i[y(t), z(t_1)]_- \rangle \\ &+ 2 \int dt_1 \varphi(t, t_1) \langle \frac{1}{2} [y(t), z(t_1)]_+ \rangle \\ &- 2 \int dt_1 \bar{M}_C(t, t_1) \langle i[z(t), y(t_1)]_- \rangle \\ &- 2 \int dt_1 \varphi_C(t, t_1) \langle \frac{1}{2} [z(t), y(t_1)]_+ \rangle \\ &+ 2 \int dt_1 \bar{M}_S(t, t_1) \langle i[z(t), x(t_1)]_- \rangle \\ &+ 2 \int dt_1 \varphi_S(t, t_1) \langle \frac{1}{2} [z(t), x(t_1)]_+ \rangle, \end{aligned} \quad (7)$$

$$\begin{aligned} \langle \dot{y}(t) \rangle + \omega_0 \langle x(t) \rangle + \Delta \langle z(t) \rangle &= -2 \int dt_1 \bar{M}(t, t_1) \langle i[x(t), z(t_1)]_- \rangle \\ &- 2 \int dt_1 \varphi(t, t_1) \langle \frac{1}{2} [x(t), z(t_1)]_+ \rangle \\ &+ 2 \int dt_1 \bar{M}_C(t, t_1) \langle i[z(t), x(t_1)]_- \rangle \\ &+ 2 \int dt_1 \varphi_C(t, t_1) \langle \frac{1}{2} [z(t), x(t_1)]_+ \rangle \end{aligned}$$

$$\begin{aligned} &+ 2 \int dt_1 \bar{M}_S(t, t_1) \langle i[z(t), y(t_1)]_- \rangle \\ &+ 2 \int dt_1 \varphi_S(t, t_1) \langle \frac{1}{2} [z(t), y(t_1)]_+ \rangle, \end{aligned} \quad (8)$$

$$\begin{aligned} \langle \dot{z}(t) \rangle - \Delta \langle y(t) \rangle &= 2 \int dt_1 \bar{M}_C(t, t_1) \langle i[x(t), y(t_1)]_- \rangle \\ &+ 2 \int dt_1 \varphi_C(t, t_1) \langle \frac{1}{2} [x(t), y(t_1)]_+ \rangle \\ &- 2 \int dt_1 \bar{M}_S(t, t_1) \langle i[x(t), x(t_1)]_- \rangle \\ &- 2 \int dt_1 \varphi_S(t, t_1) \langle \frac{1}{2} [x(t), x(t_1)]_+ \rangle \\ &- 2 \int dt_1 \bar{M}_C(t, t_1) \langle i[y(t), x(t_1)]_- \rangle \\ &- 2 \int dt_1 \varphi_C(t, t_1) \langle \frac{1}{2} [y(t), x(t_1)]_+ \rangle \\ &- 2 \int dt_1 \bar{M}_S(t, t_1) \langle i[y(t), y(t_1)]_- \rangle \\ &- 2 \int dt_1 \varphi_S(t, t_1) \langle \frac{1}{2} [y(t), y(t_1)]_+ \rangle. \end{aligned} \quad (9)$$

At this point we apply the symmetrized variant of the quantum Furutsu-Novikov theorem^{16,17}

$$\langle \frac{1}{2} [Q_i^{(0)}(t), x(t)]_+ \rangle = \int dt_1 \bar{M}_{ij}(t, t_1) \langle i[x(t), \sigma_j(t_1)]_- \rangle. \quad (10)$$

This theorem follows immediately from the fact that the spin operators $\sigma_j(t)$ are functionals of heat bath variables $\{Q_i^{(0)}\}$. An average $\langle Q_i^{(0)}(t)\sigma_j(t) \rangle$ can be found by pairing of the operator $Q_i^{(0)}(t)$ with any one operator $Q_k^{(0)}(t_1)$ involved in the functional $\sigma_j(t)$; in so doing the operator $Q_k^{(0)}(t_1)$ must be eliminated from $\sigma_j(t)$. As a result a functional derivative, or, that is the same, a commutator appears in Eq. (10). We denote here $\bar{M}(t, t_1) = M(t, t_1)\theta(t-t_1)$, $\bar{M}_C(t, t_1) = \bar{M}(t, t_1)\cos \omega_0(t-t_1)$, $\bar{M}_S(t, t_1) = \bar{M}(t, t_1)\sin \omega_0(t-t_1)$, $[A, B]_{\pm} = AB \pm BA$.

To simplify the non-Markovian equations (7)–(9) obtained we use the weak-damping approximation and suppose that the evolution of spin operators in the collision terms of Eqs. (7)–(9) is governed by the free Hamiltonian $H_0 = (\Delta/2)x$ only. It follows from free Eq. (6) with all heat bath variables $Q(t)$ being omitted that the spin operators will oscillate with the frequency $\Omega = \sqrt{\Delta^2 + \omega_0^2}$ for a retardation time $\tau = t - t_1$. The weak-damping limit which is particularly appropriate for a spin coupling to the super-Ohmic heat bath¹⁸ allows one to calculate the commutators in Eqs. (7)–(9) and to obtain the simple equations for averaged spin variables $x_1 = x, x_2 = y, x_3 = z$ at the instant t :

$$\dot{x}_i - (\Lambda_{ij} - \Gamma_{ij})x_j = \alpha_i. \quad (11)$$

We drop also the brackets $\langle \dots \rangle$ which designate averaging over the initial state of the heat bath with a temperature T . The coefficients $\alpha_i, \Gamma_{ij} (i, j = x, y, z)$ are eventually determined by the Fourier transforms of the response function (2) and covariance (3), namely, by the heat bath susceptibility $\chi_{ij}(\omega) = \delta_{ij} \chi(\omega)$, $J(\omega) = \text{Im} \chi(\omega)$, and by the spectral density $S_{ij}(\omega) = \delta_{ij} S(\omega)$ of heat bath fluctuations:

$$\alpha_1 = -2 \frac{\Delta}{\Omega} \left[J(\Omega) + \frac{\omega_0}{\Omega} J(\omega_0) + \frac{1}{2} \left(1 - \frac{\omega_0}{\Omega} \right) J(\Omega + \omega_0) + \frac{1}{2} \left(1 + \frac{\omega_0}{\Omega} \right) J(\Omega - \omega_0) \right], \quad (12)$$

$$\alpha_3 = -2 \frac{\Delta^2}{\Omega^2} J(\omega_0) - \frac{(\Omega - \omega_0)^2}{\Omega^2} J(\Omega + \omega_0) + \frac{(\Omega + \omega_0)^2}{\Omega^2} J(\Omega - \omega_0), \quad (13)$$

$$\Gamma_{11} = 2 \frac{\omega_0^2}{\Omega^2} S(0) + 2 \frac{\Delta^2}{\Omega^2} S(\Omega) + \left(1 - \frac{\omega_0}{\Omega} \right) S(\Omega + \omega_0) + \left(1 + \frac{\omega_0}{\Omega} \right) S(\Omega - \omega_0), \quad (14)$$

$$\Gamma_{22} = 2 \frac{\omega_0^2}{\Omega^2} S(0) + 2 \frac{\Delta^2}{\Omega^2} [S(\Omega) + S(\omega_0)] - \frac{\omega_0}{\Omega} \left[\left(1 - \frac{\omega_0}{\Omega} \right) S(\Omega + \omega_0) - \left(1 + \frac{\omega_0}{\Omega} \right) S(\Omega - \omega_0) \right], \quad (15)$$

$$\Gamma_{33} = 2 \frac{\Delta^2}{\Omega^2} S(\omega_0) + \frac{(\Omega - \omega_0)^2}{\Omega^2} S(\Omega + \omega_0) + \frac{(\Omega + \omega_0)^2}{\Omega^2} S(\Omega - \omega_0), \quad (16)$$

$$\Gamma_{31} = 2 \frac{\omega_0 \Delta}{\Omega^2} S(\omega_0) + \frac{\Delta}{\Omega} \left[\left(1 - \frac{\omega_0}{\Omega} \right) S(\Omega + \omega_0) - \left(1 + \frac{\omega_0}{\Omega} \right) S(\Omega - \omega_0) \right], \quad (17)$$

$$\Gamma_{13} = 2 \frac{\omega_0 \Delta}{\Omega^2} [S(0) - S(\Omega)]. \quad (18)$$

In terms of diagrammatic technique these results can be obtained if we will apply the free Green functions determined by the Hamiltonian $H_0 = (\Delta/2)x$ to calculate (anti)commutators like $\langle [x(t), z(t_1)]_{\pm} \rangle$. The designation $\text{Im} \chi(\omega) = J(\omega)$ is used here because the imaginary part $\text{Im} \chi(\omega)$ of the susceptibility (2) plays the role of the spectral function $J(\omega)$ used in dissipative quantum mechanics;¹⁸ in so doing the functions $S(\omega)$ and $J(\omega)$ are related by the Callen-Welton fluctuation-dissipation theorem: $S(\omega) = J(\omega) \coth(\omega/2T)$. It should be noted that fluctuation and dissipative characteristics of the spin subsystem subjected to the strong electromagnetic radiation do not need to be under

constraints imposed by the linear fluctuation-dissipation theorem. In this case quadratic and cubic characteristics of the our nonequilibrium system must obey the nonlinear fluctuation-dissipation relations.^{19,20} It should be stressed, however, that a compliance of the results obtained with the nonlinear fluctuation-dissipation theorem is not verified here.

The coefficients $\Gamma_{12}, \Gamma_{21}, \Gamma_{23}, \Gamma_{32}$ are not written out because they contribute only to frequency shifts which are of little interest here. We drop also the coefficient α_2 because it makes a negligible contribution to the steady-state spin projections calculated below.

For the steady-state spin projections x_0, y_0, z_0 in the rotating frame of reference we obtain from Eqs. (11)–(18) that $y_0 = 0$, and

$$x_0 = -\frac{\Delta}{\Omega} \frac{\beta(\Delta, \omega_0)}{\gamma(\Delta, \omega_0; T)}; \sigma_z^0 = z_0 = \frac{\omega_0}{\Omega} \frac{\beta(\Delta, \omega_0)}{\gamma(\Delta, \omega_0; T)}, \quad (19)$$

where

$$\beta(\Delta, \omega_0) = 2 \frac{\Delta^2}{\Omega^2} J(\Omega) + \frac{(\Omega + \omega_0)^2}{\Omega^2} J(\Omega - \omega_0) + \frac{(\Omega - \omega_0)^2}{\Omega^2} J(\Omega + \omega_0), \quad (20)$$

$$\gamma(\Delta, \omega_0; T) = 2 \frac{\Delta^2}{\Omega^2} J(\Omega) \coth\left(\frac{\Omega}{2T}\right) + \frac{(\Omega + \omega_0)^2}{\Omega^2} J(\Omega - \omega_0) \times \coth\left(\frac{\Omega - \omega_0}{2T}\right) + \frac{(\Omega - \omega_0)^2}{\Omega^2} J(\Omega + \omega_0) \times \coth\left(\frac{\Omega + \omega_0}{2T}\right), \quad (21)$$

with $\Omega = \sqrt{\Delta^2 + \omega_0^2}$.

As mentioned above, we ignore the shift of the precession frequency Ω that is due to the spin-environment interaction. Assume now that the spin involved lies in the plane (x, z) at the moment $t=0$ with the initial projections $\{x(0), 0, z(0)\}$, whereas the rotating magnetic field is switched on at the moment $t = -\infty$. Then, the free-induction decay of the spin-polarized particle is described by the following expressions:

$$x(t) = [1 - G_{xx}(t)]x_0 + G_{xx}(t)x(0) + G_{xz}(t)[z(0) - z_0], \quad (22)$$

$$z(t) = [1 - G_{zz}(t)]z_0 + G_{zz}(t)z(0) + G_{xz}(t)[x(0) - x_0], \quad (23)$$

where

$$G_{xx}(t) = (\Delta^2/\Omega^2) e^{-\gamma t} + (\omega_0^2/\Omega^2) e^{-\gamma_0 t} \cos \Omega t, \quad (24)$$

$$G_{xz}(t) = -(\omega_0 \Delta/\Omega^2) [e^{-\gamma t} - e^{-\gamma_0 t} \cos \Omega t], \quad (25)$$

$$G_{zz}(t) = (\omega_0^2/\Omega^2) e^{-\gamma t} + (\Delta^2/\Omega^2) e^{-\gamma_0 t} \cos \Omega t, \quad (26)$$

with the damping rates $\gamma = \gamma(\Delta, \omega_0; T)$ (21) and

$$\gamma_0 = 2(\omega_0^2/\Omega^2) S(0) + 2(\Delta^2/\Omega^2) S(\omega_0) + \frac{1}{2} \gamma. \quad (27)$$

It should be noted that at the state of equilibrium when the external magnetic field is constant in time ($\omega_0 = 0$) and

aligned with the x axis we obtain the well-known formulas⁵ for the transversal and longitudinal damping rates, respectively: $\gamma_0 = 2[S(0) + S(\Delta)]$, $\gamma = 4S(\Delta)$ as well for the equilibrium spin projection: $x_0 = \sigma_x^0 = -\tanh(\Delta/2T)$. Under circularly polarized light ($\omega_0 \neq 0$) an additional spin orientation z_0 (19) along the laser beam axis occurs. This polarization is caused by the direct transfer of the angular momentum from electromagnetic radiation to the spin-1/2. As is evident from Eqs. (19)–(21), photons with the reverse direction of the angular momentum (with the frequency ω_0 of opposite sign) produce the opposite in direction spin orientation. The steady-state spin projection z_0 reaches its peak

$$\sigma_z^0 = z_0 = (\omega_0 / \sqrt{\omega_0^2 + \Delta^2}) \quad (28)$$

at zero temperature and decreases as the temperature increases. In the classical case when $T \gg \Omega$ the spin polarization along the z axis is inversely related to the temperature.

We do not specify here the concrete model of the heat bath. Its spectral function is usually approximated by the expression¹⁸ $J(\omega) = A \omega^s e^{-|\omega|/\omega_c}$.

For spin-lattice relaxation the optical frequency ω_0 of the circularly polarized light may be in excess of the frequency ω_c , so that the second terms in Eqs. (20) and (21) make a major contribution to the coefficients $\beta(\Delta, \omega_0)$ and $\gamma(\Delta, \omega_0; T)$, respectively, and the light-induced spin polarization is

$$\sigma_z^0 = \tanh(\Delta^2/4\omega_0 T), \quad (29)$$

provided that $0 < \Delta \ll \omega_0, \Omega - \omega_0 \approx \Delta^2/2\omega_0$. This steady-state spin orientation establishes with a rate

$$\gamma = 4J(\Delta^2/2\omega_0) \coth(\Delta^2/4\omega_0 T). \quad (30)$$

With the supplementary proviso that $\Delta^2 \ll 4\omega_0 T$, the optically induced spin polarization is directly proportional to the intensity of the laser beam B^2 ($\Delta^2 = 4g^2\mu_0^2 B^2$) and is in

inverse proportion to the temperature T : $z_0 = \Delta^2/4\omega_0 T$. It is worth noting that the similar dependencies of the optically induced magnetization on the temperature and on the laser intensity are found by van der Ziel, Pershan, and Malstrom¹¹ in experimental studies of the inverse Faraday effect in the crystal of $\text{Eu}^{2+}:\text{CaF}_2$.

As Eq. (29) suggests, the spin orientation along the z axis tends to a saturation as the laser intensity increases. This trend was noted by Raja, Allen, and Sisk¹² which observed an inverse Faraday effect in $\text{Tb}_3\text{Ga}_5\text{O}_{12}$ crystals at room temperatures. Closer theoretical examination of the spin orientation induced by circularly polarized light in a medium without optical absorption requires a detail knowledge of the heat bath spectral function $J(\omega)$, however, this is beyond the scope of the present article.

In conclusion, we have studied the spin orientation in a rotating magnetic field produced by a circularly polarized light. We have shown that the combined effect of the rotating magnetic field and the dissipative environment on the spin degrees of freedom gives rise to the steady-state spin projections $\sigma_z^0 = z^0$ (19), (28), (29) along the laser beam axis. The occurrence of this polarization is due to the direct transfer of the angular momentum from electromagnetic radiation to the spin-1/2. There are in addition the spin components ($\sigma_x^0 = x^0 \cos \omega_0 t, \sigma_y^0 = x^0 \sin \omega_0 t$) rotating in synchronism with the external magnetic field in the plane perpendicular to the direction of light propagation. We have also considered the free-induction decay of the spin-polarized particle [see Eqs. (22)–(26)] and calculated the corresponding damping rates γ (21), (30), and γ_0 (27). It should be noted that our results for the dependencies of the spin polarization on the temperature and on the light intensity are in qualitative agreement with the experimental results^{11,12} concerning the inverse Faraday effect.

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