

Polarization dependence of spin-electric transitions in molecular exchange qubits

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(Received 6 October 2025; revised 15 December 2025; accepted 12 January 2026; published 6 February 2026)

Quasi-optical experiments are emerging as a powerful technique to probe magnetic transitions in molecular spin systems. However, the simultaneous presence of the electric- and magnetic-dipole induced transitions poses the challenge of discriminating between these two contributions. In addition, the identification of the spin-electron transitions can hardly rely on the peak intensity because of the current uncertainties on the value of the spin-electric coupling in most molecular compounds. Here, we compute the polarizations required for electric- and magnetic-dipole induced transitions through spin-Hamiltonian models of molecular spin triangles. We show that the polarization allows a clear discrimination between the two kinds of transitions. In addition, it allows one to identify the physical origin of the zero-field splitting in the ground multiplet, a debated issue with significant implications on the coherence properties of the spin qubit implemented in molecular spin triangles.

DOI: [10.1103/nfxt-sfg2](https://doi.org/10.1103/nfxt-sfg2)

I. INTRODUCTION

Spin-electric, or magneto-electric (ME), phenomena have been considered for several decades now. Following the first discussion on the possibility of piezomagnetic and ME effects [1], Dzyaloshinskii predicted their occurrence in Cr_2O_3 , relying on symmetry considerations [2]. The experimental confirmation was produced immediately thereafter [3] and successively corroborated by similar experiments [4]. Roughly at that time, electron paramagnetic resonance (EPR) experiments under static electric fields emerged as an alternative means for investigating spin-electric effects in diverse magnetic systems [5]. Today, a renaissance of such studies is taking place, revolving around the possibility of electric control of molecular spins in the context of quantum applications [6–8].

Electric fields can be used in different ways to manipulate molecular spins. One possibility is represented by the application of static fields, whose intensity is large enough to modify the eigenstates or the eigenvalues of the system, so as to modify its magnetic response [9–11]. Alternatively, baseband voltage pulses can be used for modulating the phase accumulation within spin-echo sequences in pulsed EPR experiments [12–15]. In a different approach, moderate low-frequency electric fields induce a modulation of the magnetic couplings and of the resulting magnetic response of the system [16–18]. On the other hand, weak and high-frequency electric fields

can resonantly induce transitions between unperturbed system eigenstates. In polynuclear systems, this allows one to access spin degrees of freedom that are not amenable to magnetic manipulation [19]. However, unlike the energies of the transitions between Zeeman levels, which are generally addressed in EPR experiments, those of the electric-field induced transitions are hardly tunable through the static magnetic field. This makes it more difficult to address them through cavities and calls for the use of quasi-optical free-space techniques, such as magneto far-infrared spectroscopy [20–22]. Such an experimental approach also offers the possibility to address the system with radiation of different polarizations, an opportunity that we extensively explore hereafter. Different mechanisms can, in principle, be responsible for a spin-electric coupling in molecular nanomagnets. At the spin-Hamiltonian level, the transitions between spin states induced by electric fields can result from the modulation—mediated by spin-orbit interaction—of different terms and parameters, ranging from the g tensors or single-ion anisotropy, to the antisymmetric exchange [23,24]. A mechanism that is not mediated by spin-orbit coupling consists in the modulation of isotropic exchange couplings [25,26]. This results from the fact that the relative alignment of two neighboring spins affects the charge distribution on the corresponding ions and on the interconnecting superexchange bridge. The relevance of this mechanism in molecular spin triangles based on transition-metal ions was confirmed by *ab initio* calculations [24,27–29] and by the successful interpretation of different experiments [18,19]. In the following, we thus assume that the electric field essentially affects the spin cluster through the modulation of the isotropic exchange, and derive the electric-dipole operator accordingly.

In the observation of spin-electric coupling, a particular interest is devoted to antiferromagnetic spin triangles [10,11,13,15,17,18,30–33]. There, in fact, spin frustration re-

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sults in the formation of a ground $S = 1/2$ quadruplet, whose states are defined by the total spin projection and by an additional degree of freedom (pseudospin). This is determined by the interaction that induces the zero-field splitting, which typically coincides with Dzyaloshinskii-Moriya, with inhomogeneous exchange couplings, or with a combination of the two. In all cases, an electric-field induced modulation of the exchange couplings can induce transitions between states with different values of the pseudospin and identical values of the total-spin projection. A particularly promising case is that of the undistorted triangle with Dzyaloshinskii-Moriya interaction (DMI). Here, the pseudospin can be identified with the scalar chirality [25,26,34–36], and the spin-electric manipulation can be performed within subspaces that are intrinsically protected from hyperfine interactions [37].

Here we extensively characterize both magnetic- and electric-dipole induced transitions in terms of the required field polarization. This is derived from the matrix elements of the magnetic and electric dipoles between eigenstates of prototypical spin triangles. In the presence of a well-defined quantization axis, magnetic-dipole induced transitions require circularly polarized light, as in the case of mononuclear paramagnetic systems [38]. The polarization required for electric-dipole induced transitions, instead, varies from linear to circular, depending on whether the zero-field splitting in the ground multiplet originates from the triangle distortion or on spin-orbit coupling. From this, it follows that polarization can be used to discriminate not only between magnetic- and electric-dipole induced transitions, but also between different terms in the spin Hamiltonian. The investigation is extended to different spin lengths ($s_i = 1/2, 3/2, 5/2$), and includes an analysis of relevant experimental geometries (Faraday and Voigt), defined by specific orientations of the field propagation direction relative to the spin-triangle and to the magnetic-field orientations.

The analysis of the polarization dependence is extended to all transitions between the four eigenstates belonging to the $S = 1/2$ ground quadruplet. Throughout the paper, we refer to the spin triangles characterized by the presence of DMI, inhomogeneous exchange, or a combination of the two as *chirality*, *spin sum*, and *exchange qubit*, respectively. This nomenclature is motivated by the idea that these three kinds of pseudospin can potentially be used to encode and manipulate quantum information. In particular, the eigenstates corresponding to the logical states “0” and “1” would be those labeled $|0\rangle$ and $|1\rangle$ in the following. Even though the long-term motivation of the present investigation is the coherent control of the molecule state and the implementation of quantum gates, its immediate purpose remains, more fundamentally, that of understanding the polarization properties of transitions between the lowest eigenstates of these spin systems and, more specifically, how polarization can be used to selectively address electric-field induced transitions. The results presented hereafter can be experimentally tested by continuous-wave quasi-optical spectroscopy, provided that this is performed on samples of equally oriented molecules.

The rest of the paper is organized as follows. In Sec. II, we derive the polarizations that are required to induce the magnetic- and electric-dipole transitions in different kinds of spin triangles. Section III is devoted to the derivation of the

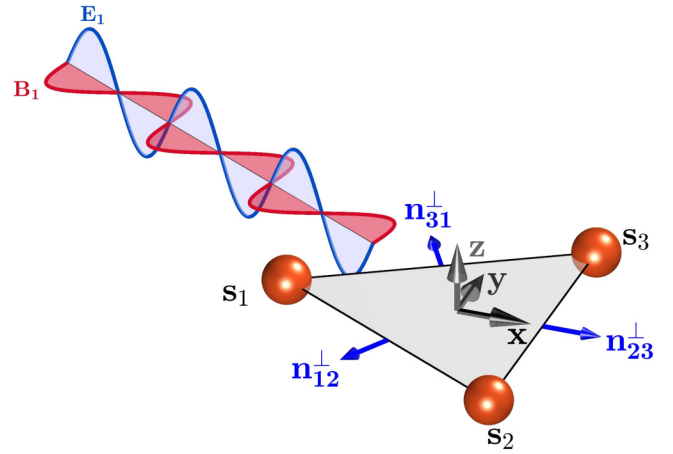


FIG. 1. Schematic view of a spin triangle interacting with a freely propagating beam. The scheme indicates the convention used in the calculations for the molecular reference frame (gray) and the interspin in-plane unit vectors $\mathbf{n}_{ij}^\perp$ (blue), perpendicular to the sides of the triangle.

polarization components that are relevant specifically in the Faraday and Voigt geometries. Our conclusions are reported in Sec. IV. Finally, in Appendix A, we explain how the required polarizations are derived from the numerical calculation of the dipole matrix elements.

II. DIPOLE OPERATORS AND MATRIX ELEMENTS

If the g tensor is isotropic and identical for all the spins within the cluster, the magnetic-dipole operator reads

$$\hat{\mu}_b = g\mu_B \hat{\mathbf{S}} \equiv \alpha_b \hat{\mathbf{S}}. \quad (1)$$

The expression of the electric-dipole operator depends on the spin-electric coupling mechanism. We consider hereafter a coupling that results from the renormalization of the exchange couplings [25–29]. In this case, one can associate an electric dipole to each of the bonds connecting two adjacent spins $\mathbf{s}_i$ and $\mathbf{s}_j$. Such dipole depends on the relative orientation of the spins and can thus be written as a vector operator that is proportional to $\mathbf{s}_i \cdot \mathbf{s}_j$ and perpendicular to the bond direction $\mathbf{r}_i - \mathbf{r}_j$, with $\mathbf{r}_i$ and $\mathbf{r}_j$ the positions of the spins.

One can always introduce a reference frame so that the three spins lie on the xy plane, at the positions $\mathbf{r}_1 = d(-1, 0)$, $\mathbf{r}_2 = \frac{d}{2}(1, -\sqrt{3})$, and $\mathbf{r}_3 = \frac{d}{2}(1, \sqrt{3})$, with d the distance between each of the two spins. The unit vectors normal to the three bonds thus are $\mathbf{n}_{12}^\perp = -\frac{1}{2}(1, \sqrt{3})$, $\mathbf{n}_{23}^\perp = (1, 0)$, and $\mathbf{n}_{31}^\perp = \frac{1}{2}(-1, \sqrt{3})$ (Fig. 1). As a result, the expression of the electric-dipole operator $\hat{\mu}_e = \alpha_e \sum_{i<j} \mathbf{n}_{ij}^\perp \cdot \hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j$ reads

$$\begin{aligned} \hat{\mu}_e = \alpha_e \mathbf{x} & \left[\hat{\mathbf{s}}_2 \cdot \hat{\mathbf{s}}_3 - \frac{1}{2}(\hat{\mathbf{s}}_1 \cdot \hat{\mathbf{s}}_2 + \hat{\mathbf{s}}_3 \cdot \hat{\mathbf{s}}_1) \right] \\ & + \alpha_e \frac{\sqrt{3}}{2} \mathbf{y} (\hat{\mathbf{s}}_3 \cdot \hat{\mathbf{s}}_1 - \hat{\mathbf{s}}_1 \cdot \hat{\mathbf{s}}_2), \end{aligned} \quad (2)$$

where α_e is the spin-electric coupling constant.

The amplitude of a dipole-induced transition $|i\rangle \rightarrow |f\rangle$ is proportional to the scalar product between the matrix element of the dipole operator $\hat{\mu}$ and the unit vector $\mathbf{e}$ that defines

the field polarization. Therefore, the polarization that maximizes the amplitude $A_{i \rightarrow f} \propto |\mathbf{e} \cdot \langle f | \hat{\mu} | i \rangle|$ corresponds to the complex conjugate of the dipole matrix element, normalized to one,

$$\mathbf{e} = \frac{\langle f | \hat{\mu} | i \rangle^*}{|\langle f | \hat{\mu} | i \rangle|} = \frac{\langle i | \hat{\mu} | f \rangle}{|\langle f | \hat{\mu} | i \rangle|}. \quad (3)$$

In the following, we compute the matrix elements of the magnetic- and electric-dipole operators between eigenstates of different spin Hamiltonians, defining chirality, partial-spin sum, and hybrid (exchange) qubits. The following analysis is limited to the ground $S = 1/2$ quadruplet. This choice is based on the fact that in the vast majority of spin triangles with direct superexchange pathways, the exchange couplings represent the dominant terms in the spin Hamiltonian, so that the coupling between the $S = 1/2$ and the $S = 3/2$ subspaces can be neglected. Moreover, it is only within the $S = 1/2$ subspace that the electric-dipole operator can induce transitions between eigenstates that are characterized by different values of some scalar spin operator (scalar chirality, partial-spin sum, or a hybrid between the two). Similar transitions cannot take place between states belonging to different S multiplets, or between states belonging to the $S = 3/2$ quadruplet. In fact, the electric-dipole operator $\hat{\mu}_e$ commutes both with $\hat{S}^2$ and with $\hat{S}_z$, and thus cannot display nonzero matrix elements between states corresponding to different values of the total spin (such as those belonging to different S multiplets) or of the total-spin projection (as is necessarily the case for two different states belonging to the $S = 3/2$ quadruplet, in a triangle formed by $1/2$ spins).

A. Dzyaloshinskii-Moriya interaction: The chirality qubit

The logical states of a chirality qubit can be identified with the states $|0\rangle$ and $|1\rangle$ of a spin triangle where the zero-field splitting in the ground quadruplet is induced by the Dzyaloshinskii-Moriya interaction. By extension, we define *chirality qubit* as a spin triangle with a Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}_c$, defined as follows [25,34–36]:

$$\hat{H}_0 = J \sum_{k=1}^3 \hat{\mathbf{s}}_k \cdot \hat{\mathbf{s}}_{k+1} + g\mu_B \mathbf{B} \cdot \hat{\mathbf{S}}, \quad (4)$$

$$\hat{H}_c = D \mathbf{z} \cdot \sum_{k=1}^3 \hat{\mathbf{s}}_k \times \hat{\mathbf{s}}_{k+1}, \quad (5)$$

with $\hat{\mathbf{s}}_4 \equiv \hat{\mathbf{s}}_1$. Here, $J > 0$ and $D > 0$ are the Heisenberg and Dzyaloshinskii-Moriya (DM) couplings, respectively. Typically, J is much larger than D , so that the four lowest eigenstates belong to the $S = 1/2$ quadruplet, on which we focus hereafter. In the case of $s_i = 1/2$ spins, the four Hamiltonian eigenstates belonging to the $S = 1/2$ subspace are also eigenstates of the scalar spin chirality,

$$\hat{C}_z = \frac{4}{\sqrt{3}} \hat{\mathbf{s}}_1 \cdot \hat{\mathbf{s}}_2 \times \hat{\mathbf{s}}_3, \quad (6)$$

with eigenvalues ± 1 (the normalization factor is included in order to have modulus one eigenvalues for $s_i = 1/2$ spins, on which we mostly focus hereafter).

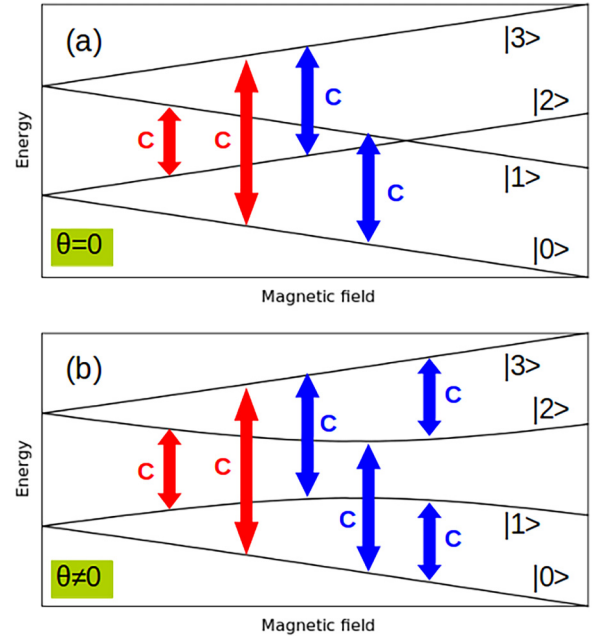


FIG. 2. Level scheme of the spin triangle with Dzyaloshinskii-Moriya interaction (chirality qubit), for magnetic field (a) oriented along the z axis ($\theta = 0$) or (b) tilted away from such axis ($\theta \neq 0$). Red and blue arrows correspond to magnetic- and electric-field induced transitions, respectively. The letter “C” denotes circular polarization.

1. Parallel magnetic field

If the magnetic field is oriented along the positive z direction, the four lowest Hamiltonian eigenstates $|C_z, S_z\rangle$, belonging to the $S = 1/2$ subspace and labeled after the values of the scalar chirality and of the total-spin projection, read [25]

$$|0\rangle \equiv | +1, -1/2 \rangle = \frac{1}{\sqrt{3}} (|\uparrow\downarrow\downarrow\rangle + \epsilon |\downarrow\uparrow\downarrow\rangle + \epsilon^* |\downarrow\downarrow\uparrow\rangle), \quad (7)$$

$$|1\rangle \equiv | -1, -1/2 \rangle = \frac{1}{\sqrt{3}} (|\uparrow\downarrow\downarrow\rangle + \epsilon^* |\downarrow\uparrow\downarrow\rangle + \epsilon |\downarrow\downarrow\uparrow\rangle), \quad (8)$$

$$|2\rangle \equiv | -1, +1/2 \rangle = \frac{1}{\sqrt{3}} (|\downarrow\uparrow\uparrow\rangle + \epsilon^* |\uparrow\downarrow\uparrow\rangle + \epsilon |\uparrow\uparrow\downarrow\rangle), \quad (9)$$

$$|3\rangle \equiv | +1, +1/2 \rangle = \frac{1}{\sqrt{3}} (|\downarrow\uparrow\uparrow\rangle + \epsilon |\uparrow\downarrow\uparrow\rangle + \epsilon^* |\uparrow\uparrow\downarrow\rangle), \quad (10)$$

where $\epsilon \equiv e^{i2\pi/3} = (-1 + i\sqrt{3})/2$. The corresponding energy levels are [Fig. 2(a)]

$$E_{0,3} = \mp \frac{1}{2} (\Delta_c + \Delta_b), \quad E_{1,2} = \mp \frac{1}{2} (\Delta_b - \Delta_c), \quad (11)$$

where $\Delta_b \equiv g\mu_B |\mathbf{B}|$ and $\Delta_c \equiv 2D$. Hereafter, we assume, for simplicity, that Δ_b is larger than Δ_c , so that E_1 is lower than E_2 and the labeling of the eigenstates reflects the ordering of the energy levels. The chirality qubit is specifically defined by two states $|0\rangle$ and $|1\rangle$ characterized by $S_z = -1/2$ and opposite values of C_z .

The nonzero matrix elements of the magnetic-dipole operator between the Hamiltonian eigenstates are

$$\langle 0|\hat{\mu}_b|0\rangle = \langle 1|\hat{\mu}_b|1\rangle = -\frac{\alpha_b}{2}\mathbf{z}, \quad (12)$$

$$\langle 3|\hat{\mu}_b|0\rangle = \langle 2|\hat{\mu}_b|1\rangle = \frac{\alpha_b}{2}(i\mathbf{y} - \mathbf{x}) \equiv -\frac{\alpha_b}{\sqrt{2}}\mathbf{n}_-, \quad (13)$$

$$\langle 3|\hat{\mu}_b|3\rangle = \langle 2|\hat{\mu}_b|2\rangle = +\frac{\alpha_b}{2}\mathbf{z}. \quad (14)$$

All other matrix elements are identically zero, as can be deduced from symmetry arguments. In fact, the total-spin components (and thus $\hat{\mu}_b$) commute with the scalar chirality operator, and therefore can only have nonzero matrix elements between states with identical values of C_z . From the above equations, it follows that the transition $|0\rangle \rightarrow |3\rangle$ (characterized by $\Delta S_z = +1$ and by a constant scalar chirality $C_z = +1$) is induced by radiation with left-handed circular polarization $\mathbf{e}_L = (\mathbf{x} + i\mathbf{y})/\sqrt{2} = \mathbf{n}_+$, with $\mathbf{e}_L$ parallel to $\langle 3|\hat{\mu}_b|0\rangle^*$. The transition $|1\rangle \rightarrow |2\rangle$ (characterized by $\Delta S_z = +1$ and by a constant scalar chirality $C_z = -1$) also requires left-handed circular polarization $\mathbf{e}_L$, which is parallel to $\langle 2|\hat{\mu}_b|1\rangle^*$.

Also the electric-field induced transitions require circular polarizations. In fact, the nonzero matrix elements of the electric dipole are

$$\langle 1|\hat{\mu}_e|0\rangle = \langle 3|\hat{\mu}_e|2\rangle^* = \frac{3\alpha_e}{4}(\mathbf{x} - i\mathbf{y}) \equiv \frac{3\alpha_e}{2\sqrt{2}}\mathbf{n}_-. \quad (15)$$

All other matrix elements are identically zero. This follows from the fact that the exchange operators $\hat{s}_i \cdot \hat{s}_j$ (and thus $\hat{\mu}_e$) commute with the total-spin projection operator, and therefore can only have nonzero matrix elements between states with equal values of S_z . The diagonal matrix elements, however, are all zero because $\langle \hat{s}_1 \cdot \hat{s}_2 \rangle = \langle \hat{s}_2 \cdot \hat{s}_3 \rangle = \langle \hat{s}_3 \cdot \hat{s}_1 \rangle = -1/4$, so that $\langle \hat{\mu}_e \rangle \propto \sum_{i<j} \mathbf{n}_{ij}^+ = 0$. From Eq. (15), it follows that the transition $|0\rangle \rightarrow |1\rangle$ ($\Delta C_z = -2$, total-spin projection $S_z = -1/2$) is induced by light with left-handed circular polarization $\mathbf{e}_L = \mathbf{n}_+$, where $\mathbf{e}_L$ is parallel to $\langle 1|\hat{\mu}_e|0\rangle^*$. The transition $|2\rangle \rightarrow |3\rangle$ ($\Delta C_z = +2$, total-spin projection $S_z = +1/2$) requires instead circular polarization $\mathbf{e}_R = \mathbf{n}_-$, with $\mathbf{e}_R$ parallel to $\langle 3|\hat{\mu}_e|2\rangle^*$.

2. Tilted magnetic field

If the magnetic field $\mathbf{B}$ lies in the xz plane and forms a nonzero angle θ with the z axis, the scalar chirality still is a good quantum number. The Hamiltonian eigenstates are also characterized by well-defined values of the spin projections $S_{\theta\pm} \equiv S_z \cos \theta_{\pm} + S_x \sin \theta_{\pm}$, where

$$\theta_{\pm} \equiv \arctan\left(\frac{\Delta_b \sin \theta}{\Delta_b \cos \theta \pm \Delta_c}\right), \quad (16)$$

and the sign in the subscript corresponds to the (positive or negative) value of C_z . The eigenstates can be expressed as follows in terms of those corresponding to the case $\theta = 0$ [Eqs. (7)–(10)]:

$$|0_{\theta}\rangle \equiv | +1, -1/2 \rangle = \cos(\theta_+/2)|0\rangle + \sin(\theta_+/2)|3\rangle, \quad (17)$$

$$|1_{\theta}\rangle \equiv | -1, -1/2 \rangle = \cos(\theta_-/2)|1\rangle + \sin(\theta_-/2)|2\rangle, \quad (18)$$

$$|2_{\theta}\rangle \equiv | -1, +1/2 \rangle = \cos(\theta_-/2)|2\rangle - \sin(\theta_-/2)|1\rangle, \quad (19)$$

$$|3_{\theta}\rangle \equiv | +1, +1/2 \rangle = \cos(\theta_+/2)|3\rangle - \sin(\theta_+/2)|0\rangle, \quad (20)$$

where the second quantum number that identifies the Hamiltonian eigenstate ($\pm 1/2$) refers to the value of $S_{\theta+}$ and $S_{\theta-}$ for positive ($|0_{\theta}\rangle$ and $|3_{\theta}\rangle$) and negative ($|2_{\theta}\rangle$ and $|1_{\theta}\rangle$) values of the scalar chirality, respectively. These two pairs of states thus form two Kramers doublets, but with two quantization axes that differ from each other and from the axis that is parallel to the static magnetic field.

The corresponding energy levels are [Fig. 2(b)]

$$E_{0,3} = \mp \frac{1}{2}[(\Delta_c + \Delta_b \cos \theta)^2 + (\Delta_b \sin \theta)^2]^{1/2}, \quad (21)$$

$$E_{1,2} = \mp \frac{1}{2}[(\Delta_c - \Delta_b \cos \theta)^2 + (\Delta_b \sin \theta)^2]^{1/2}. \quad (22)$$

Hereafter, we assume, for simplicity, that $0 \leq \theta \leq \pi/2$ and $\Delta_b \geq \Delta_c$, so that the labeling of the energy eigenvalues reflects their actual ordering.

The nonzero off-diagonal matrix elements of the magnetic-dipole operators can be derived from those obtained for $\theta = 0$ [Eqs. (12)–(14)], combined with the expressions of the eigenstates $|k_{\theta}\rangle$ [Eqs. (17)–(20)]. Their expressions read

$$\langle 3_{\theta}|\hat{\mu}_b|0_{\theta}\rangle = \frac{\alpha_b}{2}(\mathbf{z} \sin \theta_+ - \mathbf{x} \cos \theta_+ + i\mathbf{y}), \quad (23)$$

$$\langle 2_{\theta}|\hat{\mu}_b|1_{\theta}\rangle = \frac{\alpha_b}{2}(\mathbf{z} \sin \theta_- - \mathbf{x} \cos \theta_- + i\mathbf{y}). \quad (24)$$

As for the case of a parallel magnetic field, the selection rule $\Delta C_z = 0$ applies, so that all the remaining off-diagonal elements are identically zero. From the above equations, it follows that the transition $|0_{\theta}\rangle \rightarrow |3_{\theta}\rangle$ (between states with scalar chirality $C_z = +1$) is induced by light with left-handed circular polarization $\mathbf{e}_L = (\mathbf{z} \sin \theta_+ - \mathbf{x} \cos \theta_+ - i\mathbf{y})/\sqrt{2}$, propagating along the direction $\mathbf{n}_+ = \mathbf{z} \cos \theta_+ + \mathbf{x} \sin \theta_+$. The transition $|1_{\theta}\rangle \rightarrow |2_{\theta}\rangle$ (between states with scalar chirality $C_z = -1$) also requires left-handed circular polarization $\mathbf{e}_L = (\mathbf{z} \sin \theta_- - \mathbf{x} \cos \theta_- + i\mathbf{y})/\sqrt{2}$, propagating along the direction $\mathbf{n}_- = \mathbf{z} \cos \theta_- + \mathbf{x} \sin \theta_-$.

The nonzero off-diagonal matrix elements of the electric dipole are given by the expressions

$$\langle 2_{\theta}|\hat{\mu}_e|0_{\theta}\rangle = \langle 3_{\theta}|\hat{\mu}_e|1_{\theta}\rangle^* = \frac{3\alpha_e}{4}(\mathbf{x} - i\mathbf{y}) \sin(\theta_+ - \theta_-), \quad (25)$$

$$\langle 1_{\theta}|\hat{\mu}_e|0_{\theta}\rangle = \langle 3_{\theta}|\hat{\mu}_e|2_{\theta}\rangle^* = \frac{3\alpha_e}{4}(\mathbf{x} - i\mathbf{y}) \cos(\theta_+ - \theta_-). \quad (26)$$

From the above equations, it follows that independently of the orientation of the magnetic field, the circular polarization $\mathbf{e}_L$ is required for the transitions $|0_{\theta}\rangle \rightarrow |2_{\theta}\rangle$ and $|0_{\theta}\rangle \rightarrow |1_{\theta}\rangle$ and the circular polarization $\mathbf{e}_R$ is required for the transitions $|2_{\theta}\rangle \rightarrow |3_{\theta}\rangle$ and $|1_{\theta}\rangle \rightarrow |3_{\theta}\rangle$. However, the tilting angle θ and the zero-field splitting Δ_c determine the amplitudes of all these transitions, through the difference between the angles θ_+ and θ_- . In particular, a nonzero tilting angle allows the transitions $|0_{\theta}\rangle \rightarrow |2_{\theta}\rangle$ and $|1_{\theta}\rangle \rightarrow |3_{\theta}\rangle$, which are forbidden for $\theta = 0$, and at the same time reduces the amplitudes of the transitions $|0_{\theta}\rangle \rightarrow |3_{\theta}\rangle$ and $|1_{\theta}\rangle \rightarrow |2_{\theta}\rangle$. This difference between the parallel- and the tilted-field configurations results from the fact that for $\theta = 0$, the states $|0\rangle$ and $|2\rangle$ are also eigenstates of $\hat{S}_z$, corresponding to different eigenvalues

TABLE I. Summary of the polarizations required to address the electric- and magnetic-dipole induced transitions between the eigenstates ($S = 1/2$ quadruplet) of the spin triangle ($s_i = 1/2$) with Dzyaloshinskii-Moriya interaction (chirality qubit). The letters “e” and “m” denote the electric- and magnetic-dipole induced transitions, respectively. The last column indicates the polarization vector $\mathbf{e}$.

Qubit	Transition	Pol.	$\mathbf{e}$
Chirality ($\theta = 0$)	$ 0\rangle \xrightarrow{e} 1\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} + i\mathbf{y})$
	$ 0\rangle \xrightarrow{m} 3\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} + i\mathbf{y})$
	$ 1\rangle \xrightarrow{m} 2\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} + i\mathbf{y})$
	$ 2\rangle \xrightarrow{e} 3\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} - i\mathbf{y})$
Chirality ($\theta \neq 0$)	$ 0_\theta\rangle \xrightarrow{e} 1_\theta\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} + i\mathbf{y})$
	$ 0_\theta\rangle \xrightarrow{e} 2_\theta\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} + i\mathbf{y})$
	$ 0_\theta\rangle \xrightarrow{m} 3_\theta\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} \cos \theta_+ - \mathbf{z} \sin \theta_+ + i\mathbf{y})$
	$ 1_\theta\rangle \xrightarrow{m} 2_\theta\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} \cos \theta_- - \mathbf{z} \sin \theta_- + i\mathbf{y})$
	$ 1_\theta\rangle \xrightarrow{e} 3_\theta\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} - i\mathbf{y})$
	$ 2_\theta\rangle \xrightarrow{e} 3_\theta\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} - i\mathbf{y})$

($-1/2$ and $+1/2$, respectively). Therefore, the electric-dipole operator $\hat{\mu}_e$, which commutes with $\hat{S}_z$, cannot display nonzero matrix elements between the two; similarly for the states $|1\rangle$ and $|3\rangle$. For $\theta > 0$, $|0\rangle$ and $|2\rangle$ ($|1\rangle$ and $|3\rangle$) are no longer eigenstates of the same total-spin projection, but rather of the two operators $\hat{S}_{\theta+}$ and $\hat{S}_{\theta-}$, which define two different quantization axes, one for each of the chirality-related subspaces ($\mathcal{C}_z = +1$ and $\mathcal{C}_z = -1$). Therefore, the selection rules related to the conservation of the total-spin projection only apply to transitions within a given chirality-related subspace (and forbid the transitions $|0_\theta\rangle \rightarrow |3_\theta\rangle$ and $|1_\theta\rangle \rightarrow |2_\theta\rangle$), but not to intersubspace transitions.

The polarizations required for the electric- and magnetic-induced transitions are summarized in Fig. 2 and Table I. As a representative example, we apply the above equations to the case where the Zeeman splitting is twice the zero-field splitting induced by the DMI ($\Delta_b = 2\Delta_c$; Fig. 3). In particular, we plot the absorption spectra in the low-temperature limit ($k_B T \ll E_1 - E_0$), given by the function

$$J(\omega) \equiv \frac{1}{|\alpha_\chi|^2} \sum_{k=1}^3 |\mathbf{e} \cdot \langle k | \hat{\mu}_\chi | 0 \rangle|^2 e^{-(\omega - \omega_{j0})^2 / (2\sigma^2)}, \quad (27)$$

where $\omega_{jk} \equiv (E_j - E_k)/\hbar$ and σ define the central frequency and the width of the Gaussian line, respectively.

For a parallel magnetic field [$\theta = 0$; Figs. 3(a)–3(d)], the peak height related to the magnetic-dipole induced transition $|0\rangle \rightarrow |3\rangle$ (at $\hbar\omega = 3\Delta_c$) is independent of the linear polarization, and is maximal and zero for left- and right-handed polarizations, respectively. The same behavior is observed for the electric-dipole induced transition $|0\rangle \rightarrow |1\rangle$ (at $\hbar\omega = \Delta_c$). For a tilted magnetic field [$\theta = \pi/4$; Figs. 3(e)–3(h)], the electric dipole also induces the transition $|0\rangle \rightarrow |2\rangle$, while the amplitude of the $|0\rangle \rightarrow |1\rangle$ transition is reduced. The dependence on the field polarization is the same as that obtained for the parallel magnetic field.

B. Inhomogeneous exchange interaction: The partial-spin-sum qubit

The logical states of a (partial)-spin-sum qubit can be identified with the states $|0\rangle$ and $|1\rangle$ of a spin triangle where the zero-field splitting in the ground quadruplet is induced by an inhomogeneity in the exchange interaction. By extension, we define a *spin-sum qubit* as a spin triangle with a Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}_{ss}$, where $\hat{H}_0$ is given in Eq. (4) and

$$\hat{H}_{ss} = \Delta_J (\hat{\mathbf{s}}_1 \cdot \hat{\mathbf{s}}_2 + 1/4). \quad (28)$$

This term introduces an inhomogeneity between the exchange couplings. Without loss of generality, and simply to define an ordering of the eigenstates, we assume $\Delta_J > 0$. Typically, J is much larger than Δ_J , so that the four lowest eigenstates belong to the $S = 1/2$ quadruplet, on which we focus hereafter.

For all values of the spin length s_i , the four lowest eigenstates of the above spin Hamiltonian are also eigenstates of the partial-spin sum $\hat{S}_{12}^2 = (\hat{\mathbf{s}}_1 + \hat{\mathbf{s}}_2)^2$, with eigenvalues $S_{12}(S_{12} + 1)$ and $S_{12} = 0, 1$. The spin-sum qubit is specifically defined by two states characterized by the same value of the spin projection along the quantization axis and by different values of S_{12} .

1. Parallel magnetic field

If $s_i = 1/2$ and the magnetic field is oriented along the z direction, the expressions of the four lowest eigenstates $|S_{12}, S_z\rangle$, labeled by the values of the partial-spin sum and of the total-spin projection, read

$$|0\rangle \equiv |0, -1/2\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\downarrow\rangle - |\downarrow\uparrow\downarrow\rangle), \quad (29)$$

$$|1\rangle \equiv |1, -1/2\rangle = \frac{1}{\sqrt{6}}(|\uparrow\downarrow\downarrow\rangle + |\downarrow\uparrow\downarrow\rangle - 2|\downarrow\downarrow\uparrow\rangle), \quad (30)$$

$$|2\rangle \equiv |0, +1/2\rangle = \frac{1}{\sqrt{2}}(|\downarrow\uparrow\uparrow\rangle - |\uparrow\downarrow\uparrow\rangle), \quad (31)$$

$$|3\rangle \equiv |1, +1/2\rangle = \frac{1}{\sqrt{6}}(|\downarrow\uparrow\uparrow\rangle + |\uparrow\downarrow\uparrow\rangle - 2|\uparrow\uparrow\downarrow\rangle). \quad (32)$$

The corresponding energy levels are

$$E_{0,2} = -\frac{1}{2}(\Delta_J \pm \Delta_b), \quad E_{1,3} = \frac{1}{2}(\Delta_J \mp \Delta_b), \quad (33)$$

where $\Delta_B \equiv g\mu_B |\mathbf{B}|$ (Fig. 4). Hereafter, we assume, for simplicity, that Δ_J is larger than Δ_b , so that E_1 is lower than E_2 and the labeling of the eigenstates reflects the ordering of the energy levels.

The nonzero matrix elements of the magnetic-dipole operator between the Hamiltonian eigenstates are

$$\langle 0 | \hat{\mu}_b | 0 \rangle = \langle 1 | \hat{\mu}_b | 1 \rangle = -\frac{\alpha_b}{2} \mathbf{z}, \quad (34)$$

$$\langle 2 | \hat{\mu}_b | 0 \rangle = \langle 3 | \hat{\mu}_b | 1 \rangle = \frac{\alpha_b}{2} (i\mathbf{y} - \mathbf{x}) \equiv -\frac{\alpha_b}{\sqrt{2}} \mathbf{n}_-, \quad (35)$$

$$\langle 3 | \hat{\mu}_b | 3 \rangle = \langle 2 | \hat{\mu}_b | 2 \rangle = +\frac{\alpha_b}{2} \mathbf{z}. \quad (36)$$

All other matrix elements are identically zero, as can be deduced from symmetry arguments. In fact, the total-spin components (and thus $\hat{\mu}_b$) commute with the exchange operators and therefore can only have nonzero matrix elements between states with identical values of S_{12} . From the above

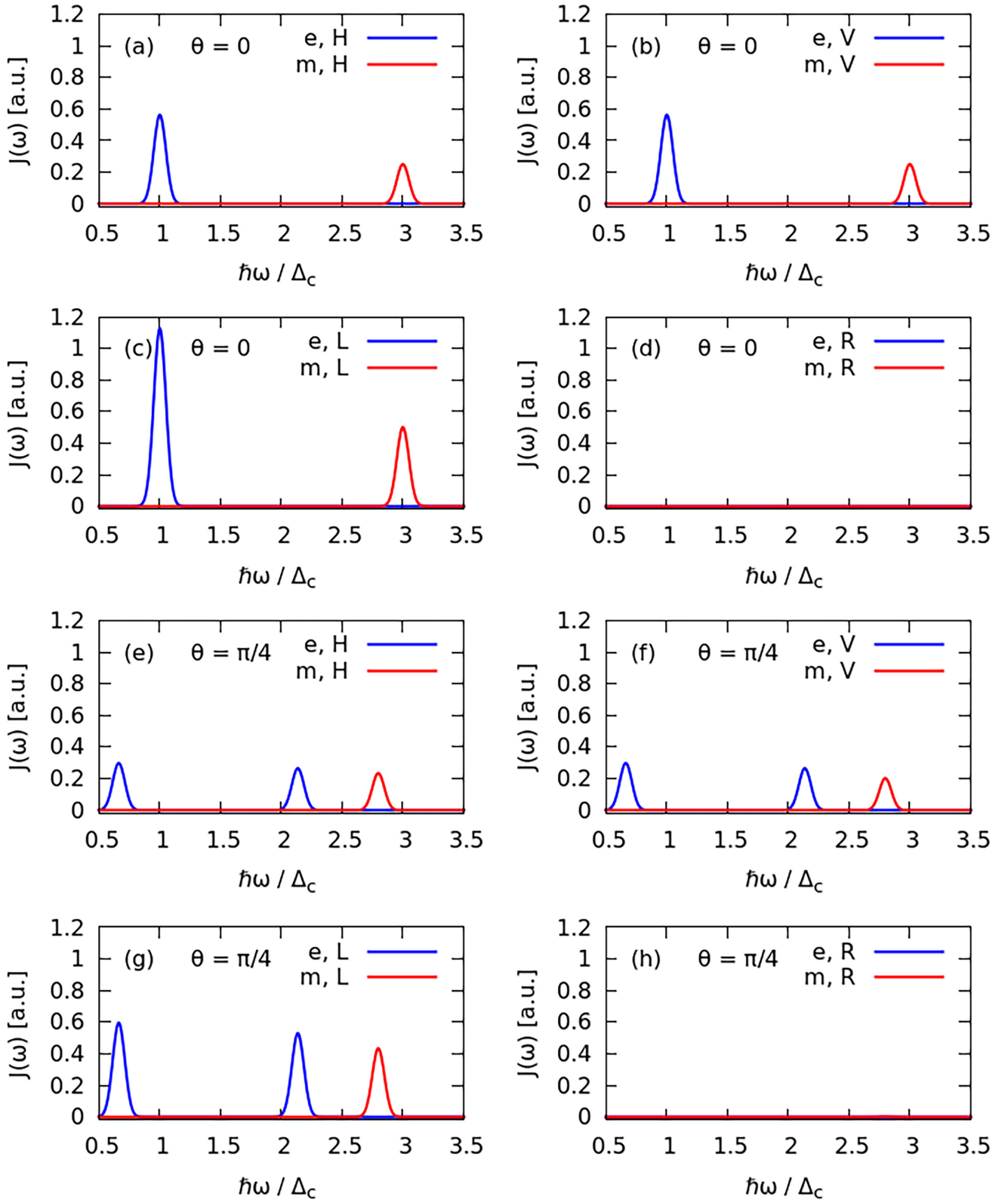


FIG. 3. Absorption spectra J [see Eq. (27)] for the spin triangle with DMI and identical exchange couplings. The Zeeman splitting is $\Delta_b = 2\Delta_c$ and the initial state is $|0\rangle$, corresponding to the low-temperature limit ($k_B T \ll \Delta_c < \Delta_b$). Blue and red lines correspond to electric- and magnetic-field induced transitions, respectively. In addition, (a)–(d) refer to the case of a longitudinal B field ($\theta = 0$), and (e)–(h) to a tilted B field ($\theta = \pi/4$). The horizontal polarizations (a), (e) H and (b), (f) V are defined by $\mathbf{e}_H = \frac{1}{2}(\mathbf{x}\sqrt{3} - \mathbf{y})$ and $\mathbf{e}_V = \frac{1}{2}(\mathbf{y}\sqrt{3} + \mathbf{x})$, respectively; the circular polarizations (c), (g) L and (d), (h) R are given by $\mathbf{e}_L$ and $\mathbf{e}_R$.

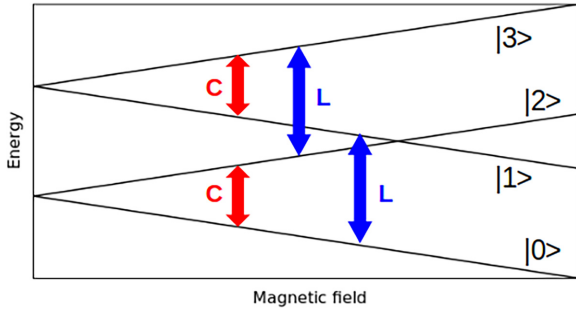


FIG. 4. Level scheme of the spin triangle with inhomogeneous Heisenberg couplings (spin-sum qubit), for arbitrary orientations of the magnetic field. Red and blue arrows correspond to magnetic- and electric-field induced transitions, respectively. The letter “C” (“L”) denotes circular (linear) polarization.

equations, it follows that both the transitions $|0\rangle \rightarrow |2\rangle$ (between states with partial spin sum $S_{12} = 0$) and $|1\rangle \rightarrow |3\rangle$ (between states with partial spin sum $S_{12} = 1$) are induced by light with left-handed circular polarization $\mathbf{e}_L = (\mathbf{x} + i\mathbf{y})/\sqrt{2}$, with $\mathbf{e}_L$ parallel to $\langle 2|\hat{\mu}_b|0\rangle^* = \langle 3|\hat{\mu}_b|1\rangle^*$.

The nonzero off-diagonal matrix elements of the electric dipole operator are

$$\langle 1|\hat{\mu}_e|0\rangle = \langle 3|\hat{\mu}_e|2\rangle = \frac{3\alpha_e}{8}(\mathbf{x}\sqrt{3} - \mathbf{y}), \quad (37)$$

and are thus orthogonal to the diagonal matrix elements, i.e., to the expectation values

$$\langle k|\hat{\mu}_e|k\rangle = -\langle k'|\hat{\mu}_e|k'\rangle = \frac{3\alpha_e}{8}(\mathbf{x} + \mathbf{y}\sqrt{3}), \quad (38)$$

where $k = 0, 2$ and $k' = 1, 3$. All other matrix elements are identically zero because the exchange operators (and thus $\hat{\mu}_e$) commute with the total spin components, and therefore can only have nonzero matrix elements between states with identical values of S_z . From Eq. (37), it follows that the transitions $|0\rangle \rightarrow |1\rangle$ and $|2\rangle \rightarrow |3\rangle$ are induced by light with linear (“Horizontal”) polarization $\mathbf{e}_H = (\mathbf{x}\sqrt{3} - \mathbf{y})/2$, with $\mathbf{e}_H$ parallel to $\langle 1|\hat{\mu}_e|0\rangle^* = \langle 3|\hat{\mu}_e|2\rangle^*$.

2. Tilted magnetic field

The zero-field Hamiltonian of the spin-sum qubit is isotropic, so that the analysis performed for the case $\mathbf{B} \parallel \mathbf{z}$ does not change substantially for different orientations of the field in the xz plane. However, we briefly report on the relevant expressions for completeness.

The eigenstates can be labeled by the values of S_{12} and of the spin projection along the field direction, $S_\theta \equiv S_z \cos \theta + S_x \sin \theta$. Their expressions read

$$|0_\theta\rangle \equiv |0, -1/2\rangle = \cos(\theta/2)|0\rangle + \sin(\theta/2)|2\rangle, \quad (39)$$

$$|1_\theta\rangle \equiv |1, -1/2\rangle = \cos(\theta/2)|1\rangle + \sin(\theta/2)|3\rangle, \quad (40)$$

$$|2_\theta\rangle \equiv |0, +1/2\rangle = \cos(\theta/2)|2\rangle - \sin(\theta/2)|0\rangle, \quad (41)$$

$$|3_\theta\rangle \equiv |1, +1/2\rangle = \cos(\theta/2)|3\rangle - \sin(\theta/2)|1\rangle, \quad (42)$$

with the same energy levels reported for $\theta = 0$ (Fig. 4).

TABLE II. Summary of the polarizations required to address the electric- and magnetic-dipole induced transitions between the eigenstates ($S = 1/2$ quadruplet) of the spin triangle ($s_i = 1/2$) with inhomogeneous Heisenberg couplings (spin-sum qubit). The letters “e,” “m,” “C,” and “L” denote electric- and magnetic-dipole induced transitions, and circular and linear polarizations, respectively.

Qubit	Transition	Pol.	$\mathbf{e}$
Spin sum ($\theta = 0$)	$ 0\rangle \xrightarrow{e} 1\rangle$	L	$\frac{1}{2}(\mathbf{x}\sqrt{3} - \mathbf{y})$
	$ 0\rangle \xrightarrow{m} 2\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} + i\mathbf{y})$
	$ 1\rangle \xrightarrow{m} 3\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} + i\mathbf{y})$
	$ 2\rangle \xrightarrow{e} 3\rangle$	L	$\frac{1}{2}(\mathbf{x}\sqrt{3} - \mathbf{y})$
Spin sum ($\theta \neq 0$)	$ 0_\theta\rangle \xrightarrow{e} 1_\theta\rangle$	L	$\frac{1}{2}(\mathbf{x}\sqrt{3} - \mathbf{y})$
	$ 0_\theta\rangle \xrightarrow{m} 2_\theta\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} \cos \theta - \mathbf{z} \sin \theta + i\mathbf{y})$
	$ 1_\theta\rangle \xrightarrow{m} 3_\theta\rangle$	C	$\frac{1}{\sqrt{2}}(\mathbf{x} \cos \theta - \mathbf{z} \sin \theta + i\mathbf{y})$
	$ 2_\theta\rangle \xrightarrow{e} 3_\theta\rangle$	L	$\frac{1}{2}(\mathbf{x}\sqrt{3} - \mathbf{y})$

The nonzero matrix elements of the magnetic-dipole operator can be obtained by combining those obtained for $\theta = 0$ [Eqs. (34)–(36)] with the expressions of the eigenstates $|k_\theta\rangle$ [Eqs. (39)–(42)]. The result is

$$\langle 2_\theta|\hat{\mu}_b|0_\theta\rangle = \langle 3_\theta|\hat{\mu}_b|1_\theta\rangle = \frac{\alpha_b}{2}(\mathbf{z} \sin \theta - \mathbf{x} \cos \theta + i\mathbf{y}). \quad (43)$$

Both the transitions $|0\rangle \rightarrow |2\rangle$ and $|1\rangle \rightarrow |3\rangle$ are thus induced by light with left-handed circular polarization, $\mathbf{e}_L = (\mathbf{x} \cos \theta - \mathbf{z} \sin \theta + i\mathbf{y})/\sqrt{2}$, propagating along the direction $\mathbf{n} = \mathbf{z} \cos \theta + \mathbf{x} \sin \theta$. Transitions between states with different values of S_{12} are forbidden, as for $\theta = 0$.

The nonzero off-diagonal matrix elements of the electric-dipole operator are

$$\langle 1_\theta|\hat{\mu}_e|0_\theta\rangle = \langle 3_\theta|\hat{\mu}_e|2_\theta\rangle = \frac{3\alpha_e}{8}(\mathbf{x}\sqrt{3} - \mathbf{y}). \quad (44)$$

The transitions $|0_\theta\rangle \rightarrow |2_\theta\rangle$ and $|1_\theta\rangle \rightarrow |3_\theta\rangle$ thus require radiation with linear polarization $\mathbf{e}_H = (\mathbf{x}\sqrt{3} - \mathbf{y})/2$, propagating along any direction $\mathbf{n}$ perpendicular to $\mathbf{e}_H$, independently of the magnetic-field orientation. Transition between states with different values of S_θ is forbidden, given that $\hat{\mu}_e$ and the total-spin projection operators commute.

The polarizations required for the electric- and magnetic-induced transitions are summarized in Fig. 4 and Table II. As a representative example, we apply the above equations to the case where the Zeeman splitting is twice the zero-field splitting induced by the inhomogeneity in the exchange interactions ($\Delta_b = 2\Delta_J$; Fig. 5). For a parallel magnetic field [$\theta = 0$; Figs. 5(a)–5(d)], the peak height related to the magnetic-dipole induced transition $|0\rangle \rightarrow |2\rangle$ (at $\hbar\omega = 2\Delta_c$) is independent of the linear polarization, and is maximal and zero for left- and right-handed polarizations, respectively. A different behavior is observed for the electric-dipole induced transition $|0\rangle \rightarrow |1\rangle$ (at $\hbar\omega = \Delta_J$) because the peak height is independent of the circular polarization (L or R) and varies from minimal to maximal, as the linear polarization is rotated in the xy plane from perpendicular to parallel, to $\mathbf{e}_H = (\mathbf{x}\sqrt{3} - \mathbf{y})/2$. For a tilted magnetic field [$\theta = \pi/4$; Figs. 5(e)–5(h)], the dependence of the electric dipole transition on

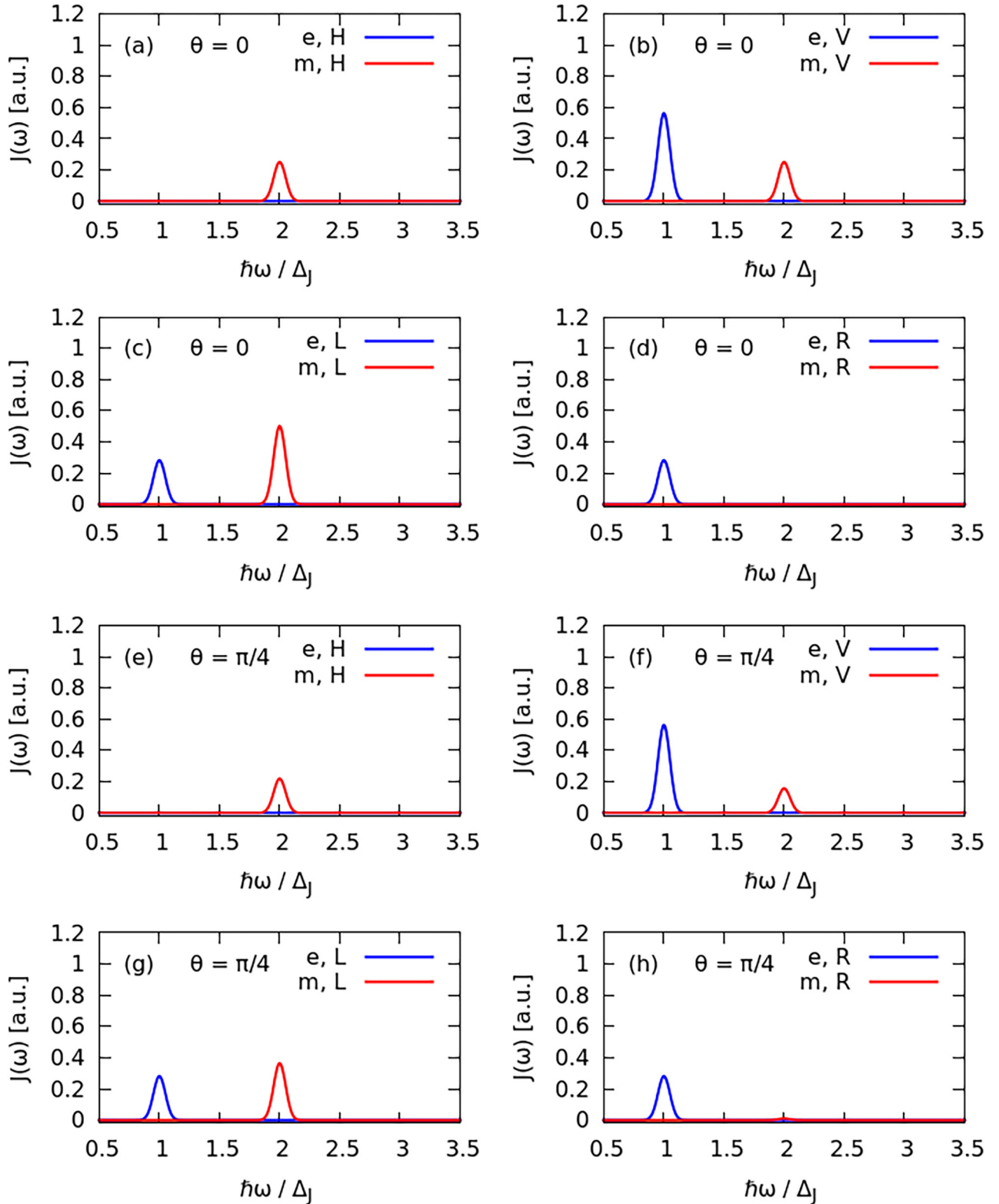


FIG. 5. Absorption spectra J [see Eq. (27)] for the spin triangle without DMI and with nonidentical exchange couplings. The Zeeman splitting is $\Delta_b = 2\Delta_J$ and the initial state is $|0\rangle$, corresponding to the low-temperature limit ($k_B T \ll \Delta_J < \Delta_b$). Blue and red lines correspond to electric- and magnetic-field induced transitions, respectively. In addition, (a)–(d) refer to the case of a longitudinal B field ($\theta = 0$), and (e)–(h) to a tilted B field ($\theta = \pi/4$). The horizontal polarizations (a), (e) H and (b), (f) V are defined by $\mathbf{e}_H = \frac{1}{2}(\mathbf{x}\sqrt{3} - \mathbf{y})$ and $\mathbf{e}_V = \frac{1}{2}(\mathbf{y}\sqrt{3} + \mathbf{x})$, respectively; the circular polarizations (c), (g) L and (d), (h) R are given by $\mathbf{e}_L$ and $\mathbf{e}_R$.

the polarization remains unchanged, while the peak heights related to magnetic-dipole transitions are consistently reduced.

3. Scalene spin triangle

The more general case where the three Heisenberg couplings are all different from one another can be dealt with, up to a term proportional to $\hat{S}^2$ (which is irrelevant within the $S = 1/2$ subspace), through the Hamiltonian term [39]

$$\hat{H}_{ss} = \Delta_J A_\varphi [\sqrt{3} \cos(2\varphi) \hat{\mathbf{s}}_1 \cdot \hat{\mathbf{s}}_2 + \sin(2\varphi) \hat{\mathbf{s}}_3 \cdot (\hat{\mathbf{s}}_1 - \hat{\mathbf{s}}_2)]. \quad (45)$$

The prefactor A_φ is defined as the inverse of the energy difference between the eigenstates of the term in square parentheses, and is such that the energy gap between the eigenstates of $\hat{H}_{ss}$ is always Δ_J , independently of φ . The angle φ defines the eigenstates through the following expressions:

$$|0_\varphi\rangle = \cos \varphi |0\rangle + \sin \varphi |1\rangle, \quad (46)$$

$$|1_\varphi\rangle = \cos \varphi |1\rangle - \sin \varphi |0\rangle, \quad (47)$$

$$|2_\varphi\rangle = \cos \varphi |2\rangle + \sin \varphi |3\rangle, \quad (48)$$

$$|3_\varphi\rangle = \cos \varphi |3\rangle - \sin \varphi |2\rangle, \quad (49)$$

where the components $|k\rangle$ are defined in Eqs. (29)–(32). The above states $|0_\varphi\rangle$ and $|2_\varphi\rangle$ ($|1_\varphi\rangle$ and $|3_\varphi\rangle$) correspond to $S_{12} = 0$ ($S_{12} = 1$) for $\varphi = 0$, to $S_{31} = 0$ ($S_{31} = 1$) for $\varphi = \pi/3$, and to $S_{23} = 0$ ($S_{23} = 1$) for $\varphi = 2\pi/3$. For generic values of φ , none of the partial-spin sums are a good quantum number. However, as discussed below, the resulting qubit, defined by a pair of states with an identical value of S_z (for example, $|0_\varphi\rangle$ and $|1_\varphi\rangle$), displays the same properties as the partial-spin-sum qubit.

In fact, the matrix elements of the magnetic dipole operator are the same as in the case $\varphi = 0$ [Eqs. (34)–(36)]. The same circular polarization ($\mathbf{e}_L$) is thus required to induce the transitions $|0_\varphi\rangle \rightarrow |2_\varphi\rangle$ and $|1_\varphi\rangle \rightarrow |3_\varphi\rangle$.

The matrix elements of the electric-dipole operator, obtained by combining Eqs. (46)–(49) with Eqs. (37) and (38), depend on the mixing angle φ through the expression

$$\langle 1_\varphi | \hat{\mu}_e | 0_\varphi \rangle = \langle 3_\varphi | \hat{\mu}_e | 2_\varphi \rangle = \frac{3\alpha_e}{4} [\mathbf{x} \cos(2\varphi + \pi/6) - \mathbf{y} \sin(2\varphi + \pi/6)]. \quad (50)$$

In all cases, the electric-dipole transitions are thus induced by a field with linear polarization, $\mathbf{e}_H = \mathbf{x} \cos(2\varphi + \pi/6) - \mathbf{y} \sin(2\varphi + \pi/6)$. This is perpendicular to all the expectation values, $\langle k_\varphi | \hat{\mu}_e | k_\varphi \rangle = \pm [\mathbf{x} \sin(2\varphi + \pi/6) + \mathbf{y} \cos(2\varphi + \pi/6)]$, where the positive and negative signs apply to $k = 0, 2$ and $k = 1, 3$, respectively.

The generalization to the case of a tilted magnetic field ($\theta \neq 0$) is obtained by combining the results of the last two sections.

C. Inhomogeneous exchange and Dzyaloshinskii-Moriya interaction: The generalized exchange qubit

The logical states of a generalized exchange qubit can be identified with the states $|0\rangle$ and $|1\rangle$ of a spin triangle

where the zero-field splitting in the ground quadruplet results from the interplay between an inhomogeneous exchange and the Dzyaloshinskii-Moriya interaction [19]. By extension, we define a *generalized exchange qubit* as a spin triangle whose Hamiltonian is given by $\hat{H} = \hat{H}_0 + \hat{H}_{ge}$, where

$$\hat{H}_{ge} = \hat{H}_{ss} \cos \phi + \hat{H}_c \sin \phi. \quad (51)$$

The angle ϕ thus provides the degree of hybridization between the chirality and the exchange components, which are recovered as special cases for $\phi = \pi/2$ and $\phi = 0$, respectively.

1. Parallel magnetic field

The eigenstates can be labeled by the values of S_z and of an additional quantum number, corresponding to the lowest (-1) and highest ($+1$) eigenvalues of H_{ge} :

$$|0_\phi\rangle = |-1, -1/2\rangle = \cos(\phi/2)|0\rangle + i \sin(\phi/2)|1\rangle, \quad (52)$$

$$|1_\phi\rangle = |+1, -1/2\rangle = i \sin(\phi/2)|0\rangle + \cos(\phi/2)|1\rangle, \quad (53)$$

$$|2_\phi\rangle = |-1, +1/2\rangle = \cos(\phi/2)|2\rangle - i \sin(\phi/2)|3\rangle, \quad (54)$$

$$|3_\phi\rangle = |+1, +1/2\rangle = -i \sin(\phi/2)|2\rangle + \cos(\phi/2)|3\rangle. \quad (55)$$

The first quantum number (± 1) that appears in the ket notation specifies the eigenvalue of $2\hat{H}_{ge}/\Delta$, with $\Delta \equiv (\Delta_J^2 + \Delta_c^2)^{1/2}$. The states $|k\rangle$ that appear on the right-hand side of the above equations are the eigenstates of the spin-sum qubit for $\theta = 0$ [Eqs. (29)–(32)] [40].

The energies corresponding to the states $|k_\phi\rangle$ are

$$E_{0,2} = -\frac{1}{2}(\Delta \pm \Delta_b), \quad E_{1,3} = \frac{1}{2}(\Delta \mp \Delta_b). \quad (56)$$

The zero-field splitting Δ is related to those of the chirality and of the spin-sum qubits by the relations $\Delta_c = \Delta \sin \phi$ and $\Delta_J = \Delta \cos \phi$.

The matrix elements of the magnetic-dipole operator are given by the expressions

$$\langle 3_\phi | \hat{\mu}_b | 1_\phi \rangle = \langle 2_\phi | \hat{\mu}_b | 0_\phi \rangle = -\frac{\alpha_b}{2} \cos \phi \mathbf{n}_-, \quad (57)$$

$$\langle 2_\phi | \hat{\mu}_b | 1_\phi \rangle = \langle 3_\phi | \hat{\mu}_b | 0_\phi \rangle = -\frac{i\alpha_b}{2} \sin \phi \mathbf{n}_-. \quad (58)$$

These transitions are thus induced by radiation with left-handed circular polarization, as for the chirality and for the spin-sum qubits. However, the selection rules related to C_z and S_{12} are removed for $0 < \phi < \pi/2$, such that all four transitions between states with opposite values of S_z are allowed.

Electric-dipole transitions are only allowed between states with identical value of S_z . The relevant matrix elements of the electric-dipole operator read

$$\langle 1_\phi | \hat{\mu}_e | 0_\phi \rangle = \frac{3\alpha_e}{8} [\mathbf{x}(\sqrt{3} - i \sin \phi) - \mathbf{y}(1 + i \sin \phi \sqrt{3})], \quad (59)$$

$$\langle 3_\phi | \hat{\mu}_e | 2_\phi \rangle = \frac{3\alpha_e}{8} [\mathbf{x}(\sqrt{3} + i \sin \phi) - \mathbf{y}(1 - i \sin \phi \sqrt{3})]. \quad (60)$$

These two transitions are induced by a field with elliptical polarization, defined by the unit vectors $\mathbf{e}_+ = \frac{\xi}{2} [\mathbf{x}(\sqrt{3} + i \sin \phi) - \mathbf{y}(1 - i \sin \phi \sqrt{3})] = \langle 1_\phi | \hat{\mu}_e | 0_\phi \rangle^* / |\langle 1_\phi | \hat{\mu}_e | 0_\phi \rangle|$ and $\mathbf{e}_- = \frac{\xi}{2} [\mathbf{x}(\sqrt{3} - i \sin \phi) - \mathbf{y}(1 + i \sin \phi \sqrt{3})] = \langle 3_\phi | \hat{\mu}_e | 2_\phi \rangle^* /$

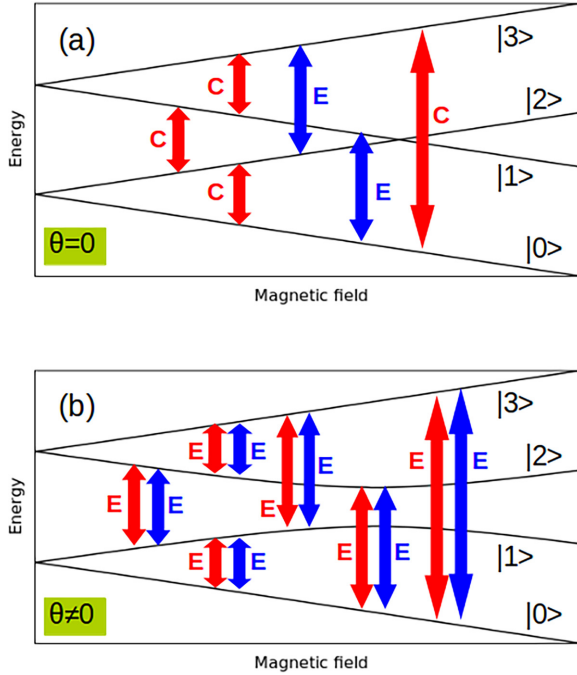


FIG. 6. Level scheme of the spin triangle with Dzyaloshinskii-Moriya interaction and inhomogeneous Heisenberg exchange (generalized exchange qubit), for magnetic field (a) oriented along the main molecule ($\theta = 0$) or (b) tilted away from such axis ($\theta \neq 0$). Red (blue) arrows correspond to magnetic-field (electric-field) induced transitions. The letters “C,” “L,” and “E” denote circular, linear, and elliptical polarizations, respectively.

$|\langle 3_\phi | \hat{\mu}_e | 2_\phi \rangle|$, where $\xi \equiv (1 + \sin^2 \phi)^{-1/2}$. From these equations, one recovers the linear polarization if the exchange inhomogeneity is much larger than the DM coupling [$\phi = 0$; Eq. (15)] and the circular polarization in the opposite limit [$\phi = \pi/2$; Eq. (37)].

The polarizations required for the electric- and magnetic-induced transitions are summarized in Fig. 6(a) and Table III.

TABLE III. Summary of the polarizations required to address the electric- and magnetic-dipole induced transitions between the eigenstates ($S = 1/2$ quadruplet) of the spin triangle ($s_i = 1/2$) with Dzyaloshinskii-Moriya interaction and inhomogeneous Heisenberg couplings (generalized exchange qubit), for a magnetic field oriented along the z axis. The letters “e” and “m” denote the electric- and magnetic-dipole induced transitions, respectively.

Qubit	Transition	Pol.	e
Exchange ($\theta = 0$)	$ 0_\phi\rangle \xrightarrow{e} 1_\phi\rangle$	E	$\frac{\xi}{2}[\mathbf{x}(\sqrt{3} + i \sin \phi) - \mathbf{y}(1 - i \sin \phi \sqrt{3})]$
	$ 0_\phi\rangle \xrightarrow{m} 2_\phi\rangle$	C	$\frac{1}{2}(\mathbf{x} + i\mathbf{y})$
	$ 0_\phi\rangle \xrightarrow{m} 3_\phi\rangle$	C	$\frac{1}{2}(\mathbf{x} + i\mathbf{y})$
	$ 1_\phi\rangle \xrightarrow{m} 2_\phi\rangle$	C	$\frac{1}{2}(\mathbf{x} + i\mathbf{y})$
	$ 1_\phi\rangle \xrightarrow{m} 3_\phi\rangle$	C	$\frac{1}{2}(\mathbf{x} + i\mathbf{y})$
	$ 2_\phi\rangle \xrightarrow{e} 3_\phi\rangle$	E	$\frac{\xi}{2}[\mathbf{x}(\sqrt{3} - i \sin \phi) - \mathbf{y}(1 + i \sin \phi \sqrt{3})]$

TABLE IV. Intensities (I_ϕ) and ellipticities (η_ϕ) of the electric- and magnetic-dipole induced transitions between the eigenstates ($S = 1/2$ quadruplet) of the spin triangle ($s_i = 1/2$) with Dzyaloshinskii-Moriya interaction and inhomogeneous Heisenberg couplings (generalized exchange qubit). The magnetic field is oriented along the z axis tilted away from the z axis ($\theta = \pi/4$).

Transition	$I_{\pi/8}$	$\eta_{\pi/8}$	$I_{\pi/4}$	$\eta_{\pi/4}$	$I_{3\pi/8}$	$\eta_{3\pi/8}$
$ 0\rangle \xrightarrow{m} 1\rangle$	0.02	0.1	0.04	0.1	0.03	0.1
$ 0\rangle \xrightarrow{e} 1\rangle$	0.6	0.3	0.8	0.6	1	0.9
$ 0\rangle \xrightarrow{m} 2\rangle$	0.4	1	0.3	0.9	0.1	0.8
$ 0\rangle \xrightarrow{e} 2\rangle$	$<10^{-2}$	0.3	$<10^{-2}$	0.6	$<10^{-2}$	0.9
$ 0\rangle \xrightarrow{m} 3\rangle$	0.05	0.8	0.2	0.8	0.4	0.9
$ 0\rangle \xrightarrow{e} 3\rangle$	0.3	0	0.08	$<10^{-2}$	0.05	0
$ 1\rangle \xrightarrow{m} 2\rangle$	0.06	0.7	0.2	0.8	0.4	0.9
$ 1\rangle \xrightarrow{e} 2\rangle$	0.05	$<10^{-2}$	0.1	$<10^{-2}$	0.07	$<10^{-2}$
$ 1\rangle \xrightarrow{m} 3\rangle$	0.4	1	0.3	0.9	0.1	0.8
$ 1\rangle \xrightarrow{e} 3\rangle$	$<10^{-2}$	0.3	$<10^{-2}$	0.6	$<10^{-2}$	0.9
$ 2\rangle \xrightarrow{m} 3\rangle$	0.02	0.1	0.04	0.1	0.03	0.1
$ 2\rangle \xrightarrow{e} 3\rangle$	0.6	0.3	0.8	0.6	1	0.9

2. Tilted magnetic field

In the presence of both exchange inhomogeneity and DM interaction ($\phi \neq 0, \pi/2$) and for tilted magnetic fields ($\theta \neq 0$), simple analytical expressions of the dipole matrix elements cannot be provided. We thus characterize the transitions in terms of their intensities, normalized to the relevant coupling constant ($\chi = e, b$),

$$I \equiv |\mathbf{e} \cdot \langle k' | \hat{\mu}_\chi | k \rangle|^2 / |\alpha_\chi|^2, \quad (61)$$

and of the degree of ellipticity, $\eta \equiv \alpha/\beta$, defined as the ratio between the major and the minor axes of the ellipse (see Appendix A). The limiting cases $\eta = 0$ and $\eta = 1$ correspond to linear and circular polarizations, respectively.

The values of I and η obtained for the generalized exchange qubit are reported in Table IV, for different values of the mixing angle ϕ . As a first remark, the interplay between the DM interaction and the inhomogeneous Heisenberg coupling, combined with the tilting of the magnetic field, removes all the selection rules that apply in the previous cases: the conservation of a scalar quantum number is not required for the transitions induced by the magnetic dipole, and the conservation of the spin projection is not required for the transitions induced by the electric dipole. Both magnetic- and electric-dipole induced transitions are thus allowed ($I \neq 0$) for each pair of eigenstates. All transitions are characterized by an elliptical polarization ($0 < \eta < 1$), even if, in some cases, the polarization is nearly linear (η is below 10^{-2}). Concerning the distinguishability between the magnetic- and the electric-dipole transitions, we note the following. The absolute values of the intensities I_ϕ cannot be used directly for this purpose because they do not include the value of the coupling constants, which are (at least in the electric case) generally unknown. However, one can rely, on the one hand, on the dependence of the intensities on the tilting angle θ and, on the other hand, on the polarization, which is always different for the electric- and magnetic-dipole induced transitions.

The polarizations required for the different transitions are summarized in Fig. 6(b). As a representative example, we apply the above equations to the case where the Zeeman splitting is twice the zero-field splitting induced by the DMI and by the inhomogeneity in the exchange interactions ($\Delta_b = 2\Delta$; Fig. 7). The magnetic dipole can induce both the $|0\rangle \rightarrow |2\rangle$ (at $\hbar\omega = 2\Delta$) and the $|0\rangle \rightarrow |3\rangle$ (at $\hbar\omega = 3\Delta$) transitions. For both values of the mixing angles [$\phi = \pi/6$, Figs. 7(a)–7(d); and $\phi = \pi/3$, Figs. 7(e)–7(h)], the height of these peaks is independent of the linear polarization, and is maximal and zero for left- and right-handed polarizations, respectively. The electric-dipole induced transition $|0\rangle \rightarrow |1\rangle$ (at $\hbar\omega = \Delta$) depends on the selected circular (L or R) or linear (H or V) polarization, approaching the behavior of the chirality qubit or of the partial-spin-sum qubit for small and large values of the mixing angle ϕ , respectively.

D. Spins larger than 1/2

We now consider triangles modeled through the same Hamiltonians $\hat{H}_c$, $\hat{H}_{ss}$, and $\hat{H}_{ge}$ considered above, but formed by larger spins, namely, $s_i = 3/2$ and $s_i = 5/2$. Systems of this kind can be implemented through Cr^{III} - and Fe^{III} -based molecules, respectively [41]. As for the triangles of $s_i = 1/2$ spins, we focus on transitions between eigenstates belonging to the ground $S \approx 1/2$ quadruplet.

One qualitative difference between the $s_i = 1/2$ and $s_i > 1/2$ cases is represented by the fact that in the latter case, in the presence of DM interaction, neither the scalar chirality C_z nor the total spin S are good quantum numbers, not even if the three exchange couplings are identical (see the Supplemental Material in Ref. [19]).

Let us start by considering the case where the magnetic field is oriented along the main symmetry axis ($\theta = 0$). As reported in Tables V and VI, all the magnetic-dipole induced transitions are induced by a circular polarization, while the electric-dipole induced transitions require a polarization that evolves from linear ($\eta = 0$) to circular ($\eta = 1$), as the strength of the DM interaction (Δ_c) increases relative to the inhomogeneity in the Heisenberg couplings (Δ_J).

When the magnetic field is tilted away from the z axis ($\theta \neq 0$) and $\Delta_J, \Delta_c \neq 0$, all transitions within the ground quadruplet can be induced both by the electric and by the magnetic components of the field, by radiation with elliptical polarizations ($0 < \eta < 1$). If either Δ_J ($\phi = \pi/2$) or Δ_c ($\phi = 0$) vanish, then electric-dipole induced transitions require linear or circular polarization, respectively, whereas magnetic-dipole induced transitions always require circular polarization (Tables V and VI). We finally note that as the spin length s_i increases, the intensities I [Eq. (61)] of the magnetic transitions display a slight increase, due to the DM-induced mixing between $S = 1/2$ and $S = 3/2$ subspaces, while those related to electric transitions undergo a much more pronounced enhancement, for all orientations of the magnetic field. This latter aspect, however, cannot be directly related to experimental data because the observed signal intensity, unlike I , also depends on the spin-electric coupling constant α_e , which can vary significantly from one molecule to the other for any given value of the spin length s_i .

TABLE V. Intensities (I_ϕ) and ellipticities (e_ϕ) of the electric- and magnetic-dipole induced transitions between the eigenstates (ground quadruplet) of the spin triangle ($s_i = 3/2$) with Dzyaloshinskii-Moriya interaction and inhomogeneous Heisenberg couplings (generalized exchange qubit).

Transition ($\theta = 0$)	I_0	η_0	$I_{\pi/4}$	$\eta_{\pi/4}$	$I_{\pi/2}$	$\eta_{\pi/2}$
$ 0\rangle \xrightarrow{e} 1\rangle$	2	0	3	0.6	5	1
$ 0\rangle \xrightarrow{m} 2\rangle$	0.5	1	0.34	1	0	–
$ 0\rangle \xrightarrow{m} 3\rangle$	0	–	0.16	1	0.5	1
$ 1\rangle \xrightarrow{m} 2\rangle$	0	–	0.16	1	0.5	1
$ 1\rangle \xrightarrow{m} 3\rangle$	0.5	1	0.34	1	0	–
$ 2\rangle \xrightarrow{e} 3\rangle$	2	$<10^{-2}$	3	0.6	5	1
Transition ($\theta = \pi/4$)	I_0	η_0	$I_{\pi/4}$	$\eta_{\pi/4}$	$I_{\pi/2}$	$\eta_{\pi/2}$
$ 0\rangle \xrightarrow{m} 1\rangle$	0	–	0.04	0.2	0	–
$ 0\rangle \xrightarrow{e} 1\rangle$	2	0	3	0.4	4	1
$ 0\rangle \xrightarrow{m} 2\rangle$	0.5	1	0.4	0.9	0	–
$ 0\rangle \xrightarrow{e} 2\rangle$	0	–	0.01	0.4	0.1	1
$ 0\rangle \xrightarrow{m} 3\rangle$	0	–	0.1	0.8	0.5	1
$ 0\rangle \xrightarrow{e} 3\rangle$	0	–	0.2	$<10^{-2}$	0	–
$ 1\rangle \xrightarrow{m} 2\rangle$	0	–	0.1	0.7	0.5	1
$ 1\rangle \xrightarrow{e} 2\rangle$	0	–	0.4	0	0	–
$ 1\rangle \xrightarrow{m} 3\rangle$	0.5	1	0.4	0.9	0	–
$ 1\rangle \xrightarrow{e} 3\rangle$	0	–	0.01	0.4	0.1	1
$ 2\rangle \xrightarrow{m} 3\rangle$	0	–	0.04	0.2	0	–
$ 2\rangle \xrightarrow{e} 3\rangle$	2.3	0*	3	0.4	4	1

TABLE VI. Intensities (I_ϕ) and ellipticities (e_ϕ) of the electric- and magnetic-dipole induced transitions between the eigenstates (ground quadruplet) of the spin triangle ($s_i = 5/2$) with Dzyaloshinskii-Moriya interaction and inhomogeneous Heisenberg couplings (generalized exchange qubit).

Transition ($\theta = 0$)	I_0	η_0	$I_{\pi/4}$	$\eta_{\pi/4}$	$I_{\pi/2}$	$\eta_{\pi/2}$
$ 0\rangle \xrightarrow{m} 2\rangle$	0.5	1	0.25	1	0	–
$ 0\rangle \xrightarrow{e} 1\rangle$	5.1	0	7.8	0.71	10	1
$ 0\rangle \xrightarrow{m} 3\rangle$	0	–	0.26	1	0.52	1
$ 2\rangle \xrightarrow{e} 1\rangle$	0	–	0.26	1	0.52	1
$ 2\rangle \xrightarrow{e} 3\rangle$	5.1	0	7.8	0.71	10	1
$ 1\rangle \xrightarrow{m} 3\rangle$	0.5	1	0.25	1	0	–
Transition ($\theta = \pi/4$)	I_0	η_0	$I_{\pi/4}$	$\eta_{\pi/4}$	$I_{\pi/2}$	$\eta_{\pi/2}$
$ 0\rangle \xrightarrow{m} 2\rangle$	0.5	1	0.3	0.9	0	–
$ 0\rangle \xrightarrow{e} 2\rangle$	0	–	0.02	0.6	0.05	1
$ 0\rangle \xrightarrow{m} 1\rangle$	0	–	0.04	0.1	0	–
$ 0\rangle \xrightarrow{e} 1\rangle$	5	$<10^{-2}$	7	0.6	10	1
$ 0\rangle \xrightarrow{m} 3\rangle$	0	–	0.2	0.8	0.5	1
$ 0\rangle \xrightarrow{e} 3\rangle$	0	–	0.7	$<10^{-2}$	0	–
$ 2\rangle \xrightarrow{m} 1\rangle$	0	–	0.2	0.8	0.5	1
$ 2\rangle \xrightarrow{e} 1\rangle$	0	–	1	$<10^{-2}$	0	–
$ 2\rangle \xrightarrow{m} 3\rangle$	0	–	0.04	0.1	0	–
$ 2\rangle \xrightarrow{e} 3\rangle$	5	$<10^{-2}$	7	0.6	10	1
$ 1\rangle \xrightarrow{m} 3\rangle$	0.5	1	0.3	0.9	0	–
$ 1\rangle \xrightarrow{e} 3\rangle$	0	–	0.02	0.6	0.05	1

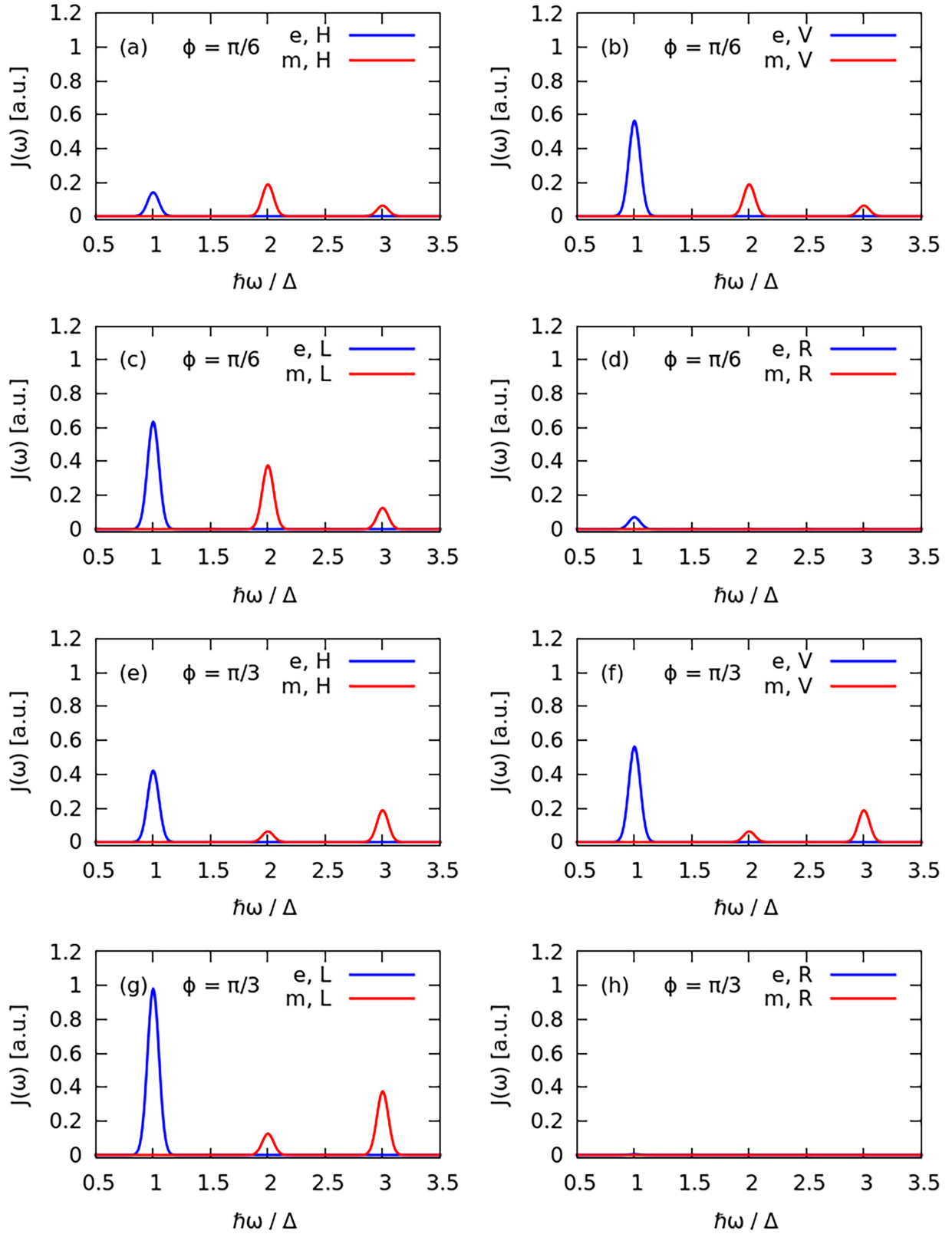


FIG. 7. Absorption spectra J [see Eq. (27)] for the spin triangle with DMI and with nonidentical exchange couplings. The Zeeman splitting is $\Delta_b = 2\Delta$, the magnetic field is longitudinal ($\theta = 0$), and the initial state is $|0\rangle$, corresponding to the low-temperature limit ($k_B T \ll \Delta < \Delta_b$). Blue and red lines correspond to electric- and magnetic-field induced transitions, respectively. In addition, (a)–(d) refer to the case $\Delta_J = \Delta_c \sqrt{3}$ ($\phi = \pi/6$), and (e)–(h) to $\Delta_c = \Delta_J \sqrt{3}$ ($\phi = \pi/3$). The horizontal polarizations (a), (e) H and (b), (f) V are defined by $\mathbf{e}_H = \frac{1}{2}(\mathbf{x}\sqrt{3} - \mathbf{y})$ and $\mathbf{e}_V = \frac{1}{2}(\mathbf{y}\sqrt{3} + \mathbf{x})$, respectively; the circular polarizations (c), (g) L and (d), (h) R are given by $\mathbf{e}_L$ and $\mathbf{e}_R$.

The spin Hamiltonians of triangles formed by spins $s_i > 1/2$ can include additional terms, such as those related to the single-ion anisotropy [42]. These terms can take different forms, depending on the (ion-dependent) orientations of the anisotropy tensor's principal axes, and on the sign and amplitude of the axial and rhombic components. A complete analysis of all of these cases is well beyond the scope of the present investigation. As a preliminary remark, we note that the presence of collinear easy- or hard-axis anisotropies was shown to scarcely affect the magneto far-infrared spectra of the Fe_3 molecule, corresponding to a triangle of $s_i = 5/2$ spins (see the Supplemental Material in Ref. [19]).

E. Typical parameter values in molecular spin triangles

Most of the above results are provided in an analytic form. They can thus be applied to any molecule that can be modeled through a spin Hamiltonian that includes three $1/2$ spins, mutually coupled by an isotropic exchange interaction and by a homogeneous DMI, and coupled to the magnetic field through an isotropic g tensor.

In general, half-integer spin triangles are given by trinuclear molecules displaying an (approximate) C_3 symmetry, and formed by half-integer spins. The length of the spin depends on the chemical element and on its oxidation state: for example, $s_i = 1/2$ spins are obtained from Cu^{II} and V^{IV} ions, $s_i = 3/2$ spins result from Cr^{III} , and $s_i = 5/2$ spins are obtained with high-spin Fe^{III} .

In known spin triangles, the identity of the metal ions, as well as that of the ligands, plays a critical role in determining the strength of the magnetic interactions. The first [25–27], but also some more recent [28,29], studies of chiral textures and spin-electric couplings within spin triangles drew values for the interaction parameters from studies on Cu_3^{II} and V_3^{IV} triangles formed within polyoxometallato (POM) supramolecular “ligands.” The large interspin distances and lack of monatomic superexchange pathways within these molecules gives rise to weak couplings, complicating the choice of spin-Hamiltonian terms and impacting the accuracy of the derived parameters. In spite of this, representative values of the J , D , and Δ_J parameters [see Eqs. (4)–(28)] can be given for this family of molecules. The very well-studied “ V_{15} ” system has been modeled as a triangle of $1/2$ spins, with $J = 1.7 \text{ cm}^{-1}$, and zero-field splitting induced either by DMI (with $D = 0.1 \text{ cm}^{-1}$ [43]) or by inhomogeneous exchange (with $\Delta_J \simeq 0.1 \text{ cm}^{-1}$ [44]), both resulting in an interdoubt gap $\Delta = 0.17 \text{ cm}^{-1}$. In a similar $(\text{V}^{\text{IV}}\text{O})_3$ POM triangle, values of the same order were estimated for the isotropic ($J = 3.3 \text{ cm}^{-1}$) and anisotropic exchanges, the latter one resulting in a zero-field splitting, $\Delta = 4.15 \times 10^{-2} \text{ cm}^{-1}$, within the quartet state [45]. The experimental data obtained with a Cu_3^{II} POM triangle were interpreted with $J \sim 3 \text{ cm}^{-1}$ and $\Delta_J, D \sim 0.3 \text{ cm}^{-1}$, resulting in a zero-field splitting, $\Delta = 0.6 \text{ cm}^{-1}$ [46].

However, the vast majority of the investigated spin triangles possess monatomic superexchange pathways, consisting of oxo, hydroxo, or alkoxo bridges. These give rise to exchange couplings at least one order of magnitude larger than those above, allowing for more precise estimates through combined magnetometric and spectroscopic studies. Although

actual values may vary broadly between molecules of the same family, a few representative examples can illustrate the typical orders of magnitude. For example, two Cu_3^{II} pyrazolato complexes have been analyzed, giving estimated values $J \sim 200 \text{ cm}^{-1}$, $\Delta_J = 24 - 63 \text{ cm}^{-1}$, and $D = 33 - 47 \text{ cm}^{-1}$; these result in zero-field splittings, $\Delta = 62 - 103 \text{ cm}^{-1}$ [47]. The estimated parameters for a similar Cu triangle are $J \sim 350 \text{ cm}^{-1}$, $\Delta_J \sim 115 \text{ cm}^{-1}$, and $D \sim 30 \text{ cm}^{-1}$, resulting in a zero-field splitting, $\Delta = 127 \text{ cm}^{-1}$ [48].

Finally, the chromium(III) and iron(III) oxo-bridged carboxylates represent the oldest and best studied families of spin triangles. Our own investigations on benzoates have yielded $J \sim 21 \text{ cm}^{-1}$, $\Delta_J \sim 0.02 \text{ cm}^{-1}$, and $D = 0.1 \text{ cm}^{-1}$ for the Cr_3^{III} complex, corresponding to a zero-field splitting, $\Delta = 0.6 \text{ cm}^{-1}$ [49]; and $J \sim 44 \text{ cm}^{-1}$, $\Delta_J = 2.2 \text{ cm}^{-1}$, and $D = 1.9 \text{ cm}^{-1}$ for the Fe_3^{III} complex, giving rise to a zero-field splitting $\Delta = 28 \text{ cm}^{-1}$ [19].

III. SPECIFIC GEOMETRIES

Hereafter, we consider the components of the dipole operators that are relevant in specific experimental geometries, defined by the direction of the light propagation $\mathbf{n}$ with respect to the static magnetic field $\mathbf{B} = B(\sin \theta, 0, \cos \theta)$. Given the propagation direction $\mathbf{n}$, the relevant part of the dipole moment for the transition $|i\rangle \rightarrow |f\rangle$ in the geometry G reads

$$\langle f|\hat{\mu}|i\rangle_G = \langle f|\hat{\mu}|i\rangle - (\mathbf{n} \cdot \langle f|\hat{\mu}|i\rangle)\mathbf{n}. \quad (62)$$

In fact, the component of the dipole that is parallel to the field-propagation direction is perpendicular to the polarization vector $\mathbf{e}$, and thus provides no contribution to the amplitude $A_{i \rightarrow f} \propto |\mathbf{e} \cdot \langle f|\hat{\mu}|i\rangle|$. The optimal polarization for this given field propagation direction is thus given by

$$\mathbf{e} = \frac{\langle i|\hat{\mu}|f\rangle_G}{|\langle f|\hat{\mu}|i\rangle_G|}. \quad (63)$$

From an opposite perspective, one might ask, given a dipole matrix element, which propagation direction maximizes the transition amplitude. The optimal $\mathbf{n}$ is the one that is orthogonal to the dipole matrix element. If $\mu_{fi} \equiv \langle f|\hat{\mu}|i\rangle$ is a real vector, or can be reduced to a real vector by applying the same phase factor to the three spatial components (i.e., if the transition is induced by a linearly polarized field), then any of the infinite directions $\mathbf{n}$ that belongs to the perpendicular plane is optimal. If, instead, the dipole matrix element is complex (circular or elliptical polarization), then one has a single optimal direction, namely, the one that is perpendicular to both the real and imaginary components of μ_{fi} .

A. Faraday geometry

In the Faraday geometry, the propagation direction is parallel to the magnetic field: $\mathbf{n} = \mathbf{n}_F \equiv (\sin \theta, 0, \cos \theta)$. From this and from Eq. (62), it follows that

$$\langle f|\hat{\mu}|i\rangle_F = \mu_{fi} - (\mu_{fi,x} \sin \theta + \mu_{fi,z} \cos \theta)\mathbf{n}_F, \quad (64)$$

with $\mu_{fi,x}$ and $\mu_{fi,z}$ the x and z components of μ_{fi} , respectively.

1. Spin triangles with Dzyaloshinskii-Moriya interaction

The component of the magnetic-dipole vector that is relevant in the Faraday geometry can be obtained by combining Eqs. (23) and (24) with the above Eq. (64). One thus obtains, for the chirality qubit, the expressions

$$\begin{aligned}\langle 3_\theta | \hat{\mu}_b | 0_\theta \rangle_F &= \frac{\alpha_b}{2} \{ \mathbf{x} [-\cos \theta_+ - \sin \theta \sin(\theta_+ - \theta)] \\ &\quad + i\mathbf{y} + \mathbf{z} [\sin \theta_+ - \cos \theta \sin(\theta_+ - \theta)] \}, \quad (65) \\ \langle 2_\theta | \hat{\mu}_b | 1_\theta \rangle_F &= \frac{\alpha_b}{2} \{ \mathbf{x} [-\cos \theta_- - \sin \theta \sin(\theta_- - \theta)] \\ &\quad + i\mathbf{y} + \mathbf{z} [\sin \theta_- - \cos \theta \sin(\theta_- - \theta)] \}. \quad (66)\end{aligned}$$

The transitions $|0_\theta\rangle \rightarrow |3_\theta\rangle$ and $|1_\theta\rangle \rightarrow |2_\theta\rangle$ can thus be induced by a field with elliptical polarization $\mathbf{e}$, given by the complex conjugates of the above transition amplitudes, normalized to 1. The ellipticity results from the fact that the imaginary (y) component of the relevant dipole matrix element coincides with that of μ_{fi} , while the real (xz) component is reduced by an amount that depends on the magnetic-field orientation.

The component of the electric-dipole vector that is relevant in the Faraday geometry can be obtained by combining Eqs. (25) and (26) with the above Eq. (64). One thus obtains, for the chirality qubit the expressions,

$$\begin{aligned}\langle 2_\theta | \hat{\mu}_e | 0_\theta \rangle_F &= \frac{3\alpha_e}{4} \sin(\theta_+ - \theta_-) \\ &\quad \times (\mathbf{x} \cos^2 \theta - i\mathbf{y} - \mathbf{z} \sin \theta \cos \theta), \quad (67) \\ \langle 1_\theta | \hat{\mu}_e | 0_\theta \rangle_F &= \frac{3\alpha_e}{4} \cos(\theta_+ - \theta_-) \\ &\quad \times (\mathbf{x} \cos^2 \theta - i\mathbf{y} - \mathbf{z} \sin \theta \cos \theta). \quad (68)\end{aligned}$$

As for the overall electric-dipole vectors, one has that $\langle 3_\theta | \hat{\mu}_e | 2_\theta \rangle_F = \langle 1_\theta | \hat{\mu}_e | 0_\theta \rangle_F^*$ and $\langle 3_\theta | \hat{\mu}_e | 1_\theta \rangle_F = \langle 2_\theta | \hat{\mu}_e | 0_\theta \rangle_F^*$. The optimal polarization for the above transitions is obtained by combining the matrix elements given in Eqs. (65)–(68) with Eq. (63).

2. Spin triangles with inhomogeneous exchange interaction

In the case of the spin-sum qubit, the polarization plane for the magnetic-dipole induced transitions is always perpendicular to the light propagation direction: $\langle f | \hat{\mu}_b | i \rangle_F = \langle f | \hat{\mu}_b | i \rangle$. The transitions are induced by a field with circular polarization, given by the normalized complex conjugates of the dipole vectors in Eq. (43).

With the electric-dipole vectors oriented on the xy plane, their relevant components in the case of a Faraday geometry read

$$\langle 1_\theta | \hat{\mu}_e | 0_\theta \rangle_F = \frac{3\alpha_e}{8} (\mathbf{x} \sqrt{3} \cos^2 \theta - \mathbf{y} - \mathbf{z} \sqrt{3} \sin \theta \cos \theta), \quad (69)$$

and $\langle 3_\theta | \hat{\mu}_e | 2_\theta \rangle_F = \langle 1_\theta | \hat{\mu}_e | 0_\theta \rangle_F$. The optimal polarization for the above transitions is obtained by combining the matrix element given in Eq. (69) with Eq. (63).

B. Voigt geometry

In the Voigt geometry, the propagation direction $\mathbf{n} = \mathbf{n}_V \equiv (\cos \theta \cos \chi, \sin \chi, -\sin \theta \cos \chi)$ is perpendicular to the magnetic field. For each direction of the magnetic field (θ), one has an infinite set of propagation directions, corresponding to different values of $0 \leq \chi < 2\pi$. From the expression of $\mathbf{n}_V$ and from Eq. (62), it follows that

$$\langle f | \hat{\mu} | i \rangle_V = \mu_{fi} - (\mu_{fi,x} \cos \theta \cos \chi + \mu_{fi,y} \sin \chi - \mu_{fi,z} \sin \theta \cos \chi) \mathbf{n}_V, \quad (70)$$

with $\mu_{fi,x}$, $\mu_{fi,y}$, and $\mu_{fi,z}$ the x , y , and z components of the dipole matrix element, respectively.

1. Spin triangles with Dzyaloshinskii-Moriya interaction

The relevant part of the magnetic-dipole operator corresponding to the transitions $|0_\theta\rangle \rightarrow |3_\theta\rangle$ and $|1_\theta\rangle \rightarrow |2_\theta\rangle$ reads

$$\begin{aligned}\langle 3_\theta | \hat{\mu}_b | 0_\theta \rangle_V &= \frac{\alpha_b}{2} [\mathbf{x}(A_+ \cos \theta \cos \chi - \cos \theta_+) \\ &\quad + \mathbf{y}(A_+ \sin \chi + i) \\ &\quad + \mathbf{z}(\sin \theta_+ - A_+ \sin \theta \cos \chi)], \quad (71)\end{aligned}$$

$$\begin{aligned}\langle 2_\theta | \hat{\mu}_b | 1_\theta \rangle_V &= \frac{\alpha_b}{2} [\mathbf{x}(A_- \cos \theta \cos \chi - \cos \theta_-) \\ &\quad + \mathbf{y}(A_- \sin \chi + i) \\ &\quad + \mathbf{z}(\sin \theta_- - A_- \sin \theta \cos \chi)], \quad (72)\end{aligned}$$

where $A_\pm \equiv \cos \chi \cos(\theta_\pm - \theta) \mp i \sin \chi$. The optimal polarization for the above transitions is obtained by combining the matrix elements given in Eqs. (71)–(72) with Eq. (63).

The relevant part of the electric dipole corresponding to the allowed transitions reads

$$\begin{aligned}\langle 1_\theta | \hat{\mu}_e | 0_\theta \rangle_V &= \frac{3\alpha_e}{4} \sin(\theta_+ - \theta_-) [\mathbf{x}(1 + B \cos \theta \cos \chi) \\ &\quad + \mathbf{y}(B \sin \chi - i) - \mathbf{z} B \sin \theta \cos \chi], \quad (73)\end{aligned}$$

$$\begin{aligned}\langle 2_\theta | \hat{\mu}_e | 0_\theta \rangle_V &= \frac{3\alpha_e}{4} \cos(\theta_+ - \theta_-) [\mathbf{x}(1 + B \cos \theta \cos \chi) \\ &\quad + \mathbf{y}(B \sin \chi - i) - \mathbf{z} B \sin \theta \cos \chi], \quad (74)\end{aligned}$$

where $B \equiv i \sin \chi - \cos \chi \cos \theta$. In addition, one has that $\langle 3_\theta | \hat{\mu}_e | 2_\theta \rangle_V = \langle 1_\theta | \hat{\mu}_e | 0_\theta \rangle_V^*$ and $\langle 3_\theta | \hat{\mu}_e | 1_\theta \rangle_V = \langle 2_\theta | \hat{\mu}_e | 0_\theta \rangle_V^*$. The optimal polarization for the above transitions is obtained by combining the matrix elements given in Eqs. (73)–(74) with Eq. (63).

2. Spin triangles with inhomogeneous exchange interaction

The relevant part of the magnetic-dipole operator for the transition $|0_\theta\rangle \rightarrow |1_\theta\rangle$ is given by

$$\begin{aligned}\langle 2_\theta | \hat{\mu}_b | 0_\theta \rangle_V &= \frac{\alpha_b}{\sqrt{2}} [\mathbf{x}(C \cos \chi - 1) \cos \theta \\ &\quad + \mathbf{y}(C \sin \chi + i) + \mathbf{z}(1 - C \cos \chi) \sin \theta], \quad (75)\end{aligned}$$

where $C \equiv \cos \chi - i \sin \chi$. In addition, $\langle 3_\theta | \hat{\mu}_b | 1_\theta \rangle_V = \langle 2_\theta | \hat{\mu}_b | 0_\theta \rangle_V$. In both cases, the transition amplitude is maximized by a field with elliptical polarization $\mathbf{e} = \langle 0_\theta | \hat{\mu}_b | 2_\theta \rangle_V / |\langle 0_\theta | \hat{\mu}_b | 2_\theta \rangle_V|$.

The relevant component of the electric-dipole vector for the transition $|0_\theta\rangle \rightarrow |1_\theta\rangle$ is given by

$$\langle 1_\theta | \hat{\mu}_e | 0_\theta \rangle_V = \frac{3\alpha_e}{8} [\mathbf{x} (D \cos \theta \cos \chi + \sqrt{3}) + \mathbf{y} (D \sin \chi - 1) - \mathbf{z} D \sin \theta \cos \chi], \quad (76)$$

where $D \equiv \sin \chi - \sqrt{3} \cos \theta \cos \chi$. In addition, $\langle 3_\theta | \hat{\mu}_e | 2_\theta \rangle_V = \langle 1_\theta | \hat{\mu}_e | 0_\theta \rangle_V$. In both cases, the transition amplitude is maximized by a field with linear polarization $\mathbf{e} = \langle 0_\theta | \hat{\mu}_b | 1_\theta \rangle_V / \langle 0_\theta | \hat{\mu}_b | 1_\theta \rangle_V$.

IV. CONCLUSIONS

We have investigated magnetic- and electric-dipole transitions in spin triangles, induced by freely propagating light beams. This exploration provides detailed tools for the interpretation of experiments carried out in the quasi-optical regime, where both the electric and the magnetic components of the oscillating field interact with the spin system. Based on the hypothesis that the electric-field induced transitions essentially result from the modulation of the symmetric exchange couplings, we have worked out the probabilities of the magnetically and electrically induced transitions for a series of spin Hamiltonians, spin lengths, polarizations, and field propagation directions relative to the magnetic field (Faraday and Voigt geometries). Overall, these results show the crucial role that the field polarization can play in identifying the physical origin of the zero-field splitting in triangular molecules and in discriminating between electric- and magnetic-field induced transitions.

More specifically, a clear difference is found between the chirality and the partial-spin-sum qubits, where the zero-field splitting is induced by Dzyaloshinskii-Moriya and by an inhomogeneous exchange, respectively. In the former case, magnetic-dipole induced transitions occur between states belonging to different doublets, the electric-dipole transitions increase from two to four as the static magnetic field passes from parallel to tilted, and the required field polarization is always circular. In the latter case, the magnetic-dipole induced transitions occur between states belonging to the same doublet, while the electric-field induced transitions are always two, induced by a linearly polarized field and independent of the magnetic field orientation and intensity. In the presence of both Dzyaloshinskii-Moriya and inhomogeneous exchange, and for a tilted magnetic field, all the selection rules resulting from the conservation of either S_z , C_z , or S_{12} break down, allowing both magnetic- and electric-dipole induced transitions between any two states belonging to the ground $S = 1/2$ quadruplet. However, different kinds of transitions are characterized by different elliptical polarizations, displaying different dependencies on the orientation of the static magnetic field. The results obtained for triangles formed by spins with length $s_i > 1/2$ qualitatively reproduce those obtained for $s_i = 1/2$, even though the matrix elements of the electric dipole grow faster with s_i than those of the magnetic dipole.

The dependence on polarization of the transition amplitudes has potential implications for the coherent control of scalar degrees of freedom (such as chirality of the partial-spin sum) in molecular spin triangles and qubits. In particular,

in a quasi-optical setup, the difference in terms of polarization between the (desired) electric-dipole transitions and the (undesired) magnetic-dipole transitions can facilitate the selective addressing of the former ones, which should otherwise rely exclusively on spectral resolution. In the presence of resonant cavities, the suppression of magnetic-dipole transitions can be achieved by placing the molecules in regions where the electric component of the field is maximal and the magnetic component is minimal. However, also in this case, polarization can be used to suppress at least the undesired transition that is spectrally closest to the target transition ($|0\rangle \leftrightarrow |1\rangle$ for qubits), thus allowing the use of shorter pulses for the implementation of quantum control and quantum gates.

ACKNOWLEDGMENTS

We acknowledge financial support from the Italian Ministero dell'Università e della Ricerca (MUR), under PNRR Project No. PE0000023-NQSTI, and from the French Agence Nationale de la Recherche (ANR), under Grant No. ANR-24-CE29-2752-01 (project SPINCHIRAL). Stimulating discussions with Milan Orlita and Jason McCoombs are also gratefully acknowledged.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX: ELLIPTICAL POLARIZATION

The derivation from the polarization vector $\mathbf{e}$ of the parameters α and β , and thus of the ellipticity η , has been carried out as follows. For a given transition $|i\rangle \rightarrow |f\rangle$, we compute the dipole vector $\mu_{fi} = \langle f | \hat{\mu} | i \rangle$ and define the relevant (optimal) polarization through the equation $\mathbf{e} = \mu_{fi}^* / |\mu_{fi}|$. The real and imaginary parts of $\mathbf{e}$ define a plane, for which we identify two mutually orthogonal vectors $\mathbf{u} \equiv \text{Re}(\mathbf{e}) / |\text{Re}(\mathbf{e})|$ and $\mathbf{v} \equiv [\text{Im}(\mathbf{e}) - [\text{Im}(\mathbf{e}) \cdot \mathbf{u}] \mathbf{u}] / [1 - (\mathbf{e} \cdot \mathbf{u})^2]$, and the corresponding components $e_u \equiv \mathbf{u} \cdot \mathbf{e}$ and $e_v \equiv \mathbf{v} \cdot \mathbf{e}$. From these, we derive four parameters, whose expressions recall those of the Stokes parameters for the electric field [50],

$$s_0 \equiv |e_u|^2 + |e_v|^2 = \alpha^2 + \beta^2, \quad (A1)$$

$$s_1 \equiv |e_u|^2 - |e_v|^2 = (\alpha^2 - \beta^2) \cos(2\gamma), \quad (A2)$$

$$s_2 \equiv 2 \text{Re}(e_u^* e_v) = (\alpha^2 - \beta^2) \sin(2\gamma), \quad (A3)$$

$$s_3 \equiv 2 \text{Im}(e_u^* e_v) = \alpha\beta, \quad (A4)$$

where α and β are the lengths of the semimajor and semiminor axes, respectively, and γ is the angle between the x and the major axis.

The validity of the above equations can be shown as follows. Denoting with $\mathbf{x} = \mathbf{u} \cos \gamma + \mathbf{v} \sin \gamma$ and $\mathbf{y} = -\mathbf{u} \sin \gamma + \mathbf{v} \cos \gamma$ the unit vectors that are parallel to the major and minor axes of the ellipse, such that $\mathbf{e} = \alpha \mathbf{x} + i\beta \mathbf{y}$, the polarization becomes $\mathbf{e} = e_u \mathbf{u} + e_v \mathbf{v}$,

where

$$e_u = \alpha \cos \gamma - i\beta \sin \gamma, \quad e_v = \alpha \sin \gamma + i\beta \cos \gamma. \quad (\text{A5})$$

Equations (A1)–(A4) follow from the above expressions of e_u and e_v in terms of α and β .

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