Free Induction Decay and Spin Echo Signals from Spin Triplet States of Axially Asymmetric Objects in Single Crystals in Zero Constant Magnetic Field: Application of Single Transition Operators

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Abstract—Anisotropic dynamics of the spin triplet states (STSs) in single crystals with the zero field splitting (ZFS) of their levels by the axially asymmetric Hamiltonian is investigated in zero constant magnetic field (ZF) under the action of the canonically oriented varying magnetic fields. The equations of motion for single transition operators (STOs) corresponding to the definite transition of ZFS are derived. The obtained equations written in terms of one averaged equation for STO vector appeared to be a particular case (for STS) of the universal equation of Feynman et al, which is valid for any kind of perturbation affecting only two levels of any quantum mechanical system. As well as that, our equation is analogous to the Bloch equation without decay for the usual magnetization components of the Zeeman system in a constant magnetic field and a transverse to it varying field. However, the motion of the observable macroscopic sample magnetization, which follows from our equations, has quite different character than that of the motion of the STO vector. Here, in terms of this magnetization the signals of the free induction decay and of the two-pulse spin echo are calculated in ZF.

Index Terms—Free Induction Decay, Single Transition Operators, Spin Echo, Spin Triplet States, Zero Constant Magnetic Field.

I. INTRODUCTION

A lot of axially asymmetric physical objects in single crystals have spin triplet states (STSs), i.e. the states with their spin equal to unity and with the three possible energy levels in zero constant magnetic field (ZF), given by the Hamiltonian of the quadrupolar form with \( E \neq 0 \) (for notations see further):

\[
\mathcal{H}_Q = D \left( S_x^2 - \frac{1}{3} S(S+1) \right) + E \left( S_x^2 - S_y^2 \right) = -X S_x^2 - Y S_y^2 - Z S_z^2.
\]

As the examples of such objects, the following ones can be mentioned: triplet excitons in photoexcited inorganic crystals (for instance, in NaNO\(_3\) [1]); some organic molecules with the triplet ground state (for instance, diphenylmethylen in benzophenone [2]; photoexcited molecules of chlorophyll and carotenoid, which are important for photosynthesis [3]; nuclei with spin 1 in the case of the axially asymmetric gradient of the electric field on them (for instance, \(^{14}\)N nuclei in cocaine [4]); the axially asymmetric guest molecules in organic crystals, acquiring STSs at the photoexcitation [5], [6]. All these physical objects find wide application in science and technique, especially the last ones [6]. Therefore, we shall concentrate our attention on these samples. At that, the pulse versions of EPR, in particular, the free induction decay (FID) experiments [6]-[8] are the most actual tools for the investigation of the dynamics of STS in ZF. In addition, spin echo experiments in ZF are employed for the STS investigation [9], [10]. Therefore, we suppose primarily to investigate the nonstationary magnetic dynamics of STSs with the zero field splitting (ZFS) of their energy levels. At that, as the result of \( E \neq 0 \) there are no degenerated quadrupolar levels in the energy spectrum of such STSs. Consequently, the approach used in [11], [12] for the description of the nonstationary nuclear quadrupole resonance in the case of the twice-degenerated quadrupolar levels with the corresponding physical interpretation of this approach is not applicable here. As it was mentioned in [9], in ZF the absence of the magnetic moment associated with any of triplet spin states is the paradox of EPR experiments under such conditions. In the given paper, we solve this paradox for the case \( E \neq 0 \) with the help of the deriving of the equations of motion for the single transition operators (STOs) determined in the basis of the usual triplet wave functions. The formalism of STOs (the concise synonym of "operators of fictitious spin 1/2") was elaborated in the series of papers [13]-[15] and is described in the monograph [16]. This formalism was applied by the authors of [13]-[15] to the investigation of the spin dynamics of nuclei with the spin 1 and with the diagonal axially symmetric quadrupolar interaction (corresponds to \( E = 0 \) in (1)), subjected to the strong constant magnetic field, which lifted the degeneration of the quadrupolar levels. At that, it was the density matrix \( \rho(t) \), which contained the time dependence at the spin evolution. Contrary to them, we suppose to investigate the spin dynamics of STS in ZF in the case of the axial asymmetry \( (E \neq 0) \), which lifts the degeneration of the quadrupolar levels. At that, the obtaining i) of the equations of motion for STOs in zero constant and nonzero varying magnetic fields; ii) of the solutions of these equations,
containing the time dependence; iii) of the observable values of the components of the sample full magnetization – is our aim.

For this purpose we elaborate first the method of the diagonalization of ZFS Hamiltonian in the basis of the usual triplet wave functions; the Hamiltonian of the spin interaction with a canonically oriented varying magnetic field is transformed correspondingly (Section II). The equations of motion for the STOs, derived with these two transformed Hamiltonians, and the exact solution of these equations in the rotating frame are presented in Section III. At that, the motion of the observable macroscopic sample magnetization, which follows from these solutions, has quite different character than that of the motion of the STO vector. For some cases of ZF EPR, this character was described in our recent paper [17]. Here, we would like to show how this motion leads to the nonzero signal of the free induction decay (FID) and that of the two-pulse spin echo. The values of these signals are calculated in Section IV. The FID signal at the absence of rotation and the steady state dynamic susceptibilities at the absence of saturation are calculated in Section V. The possibility of the nonequilibrium form of the density matrix after the photoexcitation is taken into account for all the problems considered.

II. DIAGONALIZATION OF THE ZFS HAMILTONIAN IN TERMS OF STOS

The anisotropic evolution of STS of the photoexcited axially asymmetric guest molecules in organic single crystals (for the definiteness we shall have in mind such samples, though the main following results are valid for all the objects mentioned in the Introduction) in the zero constant magnetic field under the action of the varying magnetic field is governed by the following Hamiltonian:

$$\mathcal{H} = \mathcal{H}_0^Q + \mathcal{H}'$$

(2)

where $\mathcal{H}_0^Q$ of the form (1) is the ZFS Hamiltonian (the principal Hamiltonian of STS in ZF) with the level energies $X = D / 3 - E$; $Y = D / 3 + E$; $Z = -(2/3)D$; $D, E$ are the nonzero parameters of ZFS, which everywhere in the given paper is suggested to be well resolved; $S_{X,Y,Z}$ are the projections of the full electron spin $S=1$ of STS onto the $X,Y,Z$ axes – the main axes of the tensor of the ZFS interaction $\mathcal{H}_0^Q$ (it is supposed that these axes coincide with the main axes of the $g$-factor tensor); $X,Y,Z$ axes are connected with the definite axes of molecules, possessing ZFS. The $Z$ axis is the distinguished axis of a molecule with the STS, with which the parameter $D$ is connected (see [3], [5], [6], [18]). At that, the dipole-dipole interaction of the unpaired electrons on the excited triplet molecular orbital is the cause of the ZFS described by the Hamiltonian (1) (see [3], [5]). The perturbation $\mathcal{H}' = 2g_{K}\mu_{B}B_{K}S_{K}\cos\alpha$ is the interaction of STS with the varying magnetic field $2B_{K}\cos\alpha$, polarized along the molecular axis $K = X,Y,Z$ (the so-called canonical orientations [10]); $g_{K}$ are the diagonal components of the $g$-factor tensor of STS; $\mu_{B}$ is the Bohr magneton. Until the introduction of the average (over the spin ensemble) components of the sample magnetization, our consideration is single-particle.

For the solution of the stated problems of the spin dynamics, we diagonalize the principal Hamiltonian (1) by means of the unitary transformation (UT) with the help of the operator $U_0$ presented by the matrix

$$\begin{pmatrix}
1 & 0 & 1 \\
-1 & 0 & -i \\
i & 0 & -i \\
0 & \sqrt{2} & 0
\end{pmatrix}.$$  As a result, the matrix $|T_{i+}\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$, $|T_{0}\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$, $|T_{i-}\rangle = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$ of the UT Hamiltonian is $\mathcal{H}'_0 = U_0 \mathcal{H}_0 U_0^{-1}$ appears to be diagonal in the presentation of the usual triplet spin functions $|T_{i+}\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$, $|T_{0}\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$, $|T_{i-}\rangle = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$. This fact enables us to use the described in [16] formalism of the single transition operators (fictitious spin 1/2 operators) $S_{X,Y,Z}^{i,j}$, which are determined in the same presentation. $S_{X,Y,Z}^{i,j}$ is the operator of the $X,Y,Z$-th component, respectively, of the STS spin related to the transition between the i and j levels; $i, j = X,Y,Z$ are the notations of the STS levels with the energies $X,Y,Z$ correspondingly. It should be noted that exactly the same values of the diagonal elements has the initial Hamiltonian $\mathcal{H}_0^Q$ in the presentation of its eigenfunctions $|T_{i+}\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$, $|T_{0}\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$, $|T_{i-}\rangle = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$ (see [5]).

The UT operators of the components of the full STS spin and that of the full sample magnetization are equal respectively to

$$\begin{pmatrix}
1 & 0 & 1 \\
-1 & 0 & -i \\
i & 0 & -i \\
0 & \sqrt{2} & 0
\end{pmatrix}.$$  

\footnote{The matrix presentation of the unitary operator $U_0$ introduced here supposes that the energy levels of STS satisfy the inequality $X > Y > Z$. At the rearrangement of the energy levels $i \leftrightarrow j$ in this inequality, it is necessary to rearrange the lines $i \leftrightarrow j$ in the matrix of the operator $U_0$ and the columns $i \leftrightarrow j$ in the matrix of the operator $U_0^{-1}$. For the example, see Appendix. Further, one should work with the new operators $U_0$ and $U_0^{-1}$ just in the same manner, as it is described in the following text.}

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where \( n \) is the STS concentration. In all the states \([I_{x}, I_{y}],[T_{x}],[I_{z}] \), STS has no magnetic moment in ZF [18], which fact, however, does not hinder from the magnetic resonance observation on the separate STS transitions (see below). The UT full Hamiltonian is equal to \( \mathcal{H} = \mathcal{H}_0 + \mathcal{H}^{\text{ext}} \), where the UT principal Hamiltonian

\[
\mathcal{H}_0 = 2XS_{x}^{y-z} - 2YS_{y}^{z-x} - 2ZS_{z}^{x-y}
\]

gives the STS energy levels \( X, Y, Z \); the UT with the help of (3) perturbation (interaction with the varying field polarized along the \( K \) axes) \( \mathcal{H}^{\text{ext}} = 4g_{K}\mu_{B}B_{0}S_{z}^{y} \cos \omega t \) with the indices \( K, i, j = X, Y, Z = Y, Z, X = Z, X, Y \) causes transitions between the \( i,j \) levels (see below in detail). Further, the text will be presented both in terms of the STOs and in terms of the observable (average over spin ensemble) components of the sample magnetization \( \langle M_{K}(t) \rangle = \text{Sp} \langle \rho M_{K}(t) \rangle \) (see below in detail). The equations of motion (2)-(4) and (7) in [17] were written just for the values \( \langle M_{K}(t) \rangle \). Here \( \rho \) is the diagonal density matrix describing the state of the spin system (equilibrium or reached because of the photoexcitation, as in [19], [7]-[9], respectively). At that, \( \rho \) has the population probabilities \( P_{X}, P_{Y}, P_{Z} \) of the levels \( X, Y, Z \) on its diagonal. Taking the opportunity, we would like to note that under such conditions the average value of the STS principal Hamiltonian is equal to

\[
\langle \mathcal{H}_0 \rangle = \text{Sp} \langle \rho \mathcal{H}_0 \rangle = 2X \cdot \langle S_{x}^{y-z} \rangle - 2Z \cdot \langle S_{y}^{z-x} \rangle = X \cdot P_{X} + Y \cdot P_{Y} + Z \cdot P_{Z},
\]

where \( \langle S_{z}^{y} \rangle = \text{Sp} \langle \rho S_{z}^{y} \rangle = (1/2)(P_{X} - P_{Y}) \), \( P_{X} + P_{Y} + P_{Z} = 1 \).

III. EQUATIONS OF MOTION FOR THE STOS IN ZF AND THEIR EXACT SOLUTION

Our aim, first, is to investigate the STO motion under the action of the varying field polarized along the \( K \) axis and having the frequency \( \omega \) near the absolute value of the frequency \( \omega_{0} \) near the absolute value of the frequency \( \omega_{0} \equiv (E_{z} - E_{y})h^{-1} \) of one of the STS transitions, suggested to be well resolved. They are equal to:

\[
\omega_{0}^{x-z} \equiv (Y - Z)h^{-1} = (D + E)h^{-1};
\]
\[
\omega_{0}^{z-x} \equiv (Z - X)h^{-1} = (-D + E)h^{-1};
\]
\[
\omega_{0}^{x-y} \equiv (X - Y)h^{-1} = -2Eh^{-1}.
\]

Using the quantum operator equations \( S_{x,y,z}^{\text{ext}} = (i\hbar^{-1}) [S_{x,y,z}^{\text{ext}}, \mathcal{H}] \) and the commutation relations for STOs [16], the following exact equations of the forced evolution of these operators can be easily obtained:

\[
\dot{S}_{x}^{i,j} = -\omega_{0}^{i,j}S_{x}^{i,j} + 4g_{K}\mu_{B}B_{0}^{i,j}S_{z}^{y} \cos \omega t
\]
\[
\dot{S}_{y}^{i,j} = -\omega_{0}^{i,j}S_{y}^{i,j} + 4g_{K}\mu_{B}B_{0}^{i,j}S_{z}^{y} \cos \omega t
\]
\[
\dot{S}_{z}^{i,j} = -4g_{K}\mu_{B}B_{0}^{i,j} \cos \omega t,
\]

where \( \omega_{0}^{i,j} = g_{K}^{i,j}\mu_{B}h^{-1}B_{0}^{i,j} \). With \( K, i, j = X, Y, Z = Y, Z, X = Z, X, Y \). At that, the only nonzero component of the sample magnetization (average over spin ensemble), created as a result of this forced evolution, is equal to \( \langle \dot{S}_{x}^{i,j}(t) \rangle = \text{Sp} \langle \rho M_{K}(t) \rangle \), where

\[
M_{K}(t) = -2ng_{K}^{i,j}\mu_{B}S_{z}^{y}(t)\text{ with the same set of indices } K, i, j.
\]

The time dependence of \( M_{K}(t) \) operators follows from the solution of the operator equations of motion (7) for the component \( S_{x}^{i,j}(t) \) of the operator of the single transition vector \( S_{x}^{i,j}(t) \) subjected to the action of the full Hamiltonian \( \mathcal{H}^{\text{ext}} \). We would like to note that, as one would expect, (7) does not follow from (3) of [20] at the zero value of the constant magnetic field. As it is seen from (7), EPR at the frequency of the varying field \( \omega \approx \omega_{0}^{i,j} \) can be excited and registered in single crystals by the receiver/transmitter coil (as in [7], [8]), oriented along the \( K \) axis, what is noted by the indices at the values \( \omega_{0}^{i,j}, g_{K}^{i,j}, B_{0}^{i,j}, M_{K}(t) \) in (7) and after it (not to be confused with the indices of STOs \( S_{x,y,z}^{\text{ext}} \)).

It follows from (7) that at the absence of a varying field each single transition vector \( \vec{S}_{x}^{i,j}(t) \) with the components \( \langle S_{x}^{i,j}(t), S_{y}^{i,j}(t), S_{z}^{i,j}(t) \rangle \) accomplishes precession with the \( \omega_{0}^{i,j} \) frequency around the \( Z \) axis at the constant value of \( S_{z}^{y} \) (of course, this precession is to be excited, what is realized by the authors of [7], [8] by the pulse of the varying field). Equations (7) with the presence of a varying field, valid for STOs without decay in the laboratory frame (LF), are the analogue of the Bloch equations without decay for the magnetization components of a Zeeman spin system in LF. It is seen from here and from the further calculations that the motion of the single transition vector \( \vec{S}_{x}^{i,j}(t) \) under the action of the varying field \( 2B_{0}^{i,j} \cos \omega t \) is analogous to the motion of the magnetization vector of the usual Zeeman.
spin system in a constant magnetic field and a transverse to it varying field. This similarity was mentioned also in [9], [10]. As expected, (7) have the form of the universal equation of motion \( \vec{\tau} = \left[ \vec{a} \times \vec{\tau} \right] \) of the vector \( \vec{\tau} \) in the abstract linear vector space [21], which is valid for any kind of perturbation affecting only two levels of any quantum mechanical system. In [10], dedicated to the STS in ZF, in the case \( E \neq 0 \), the \( \vec{a} \) vector in the equation \( \vec{\tau} = \left[ \vec{a} \times \vec{\tau} \right] \), written for \( Z - Y \) transition, has the components \((0,4\alpha_{XY}^0 \cos \omega t, \alpha_{YY}^0 - \omega^2)\). Using the STO definitions [16], it is easy to notice (see Appendix) that the vector \( \vec{\tau} \) in our notations and with the sequence of the axes and levels of \([10]\), differing from our sequence, has the components \( \{2Sp\{\rho(t)S_{X}^{Z-Y}\}, 2Sp\{\rho(t)S_{Y}^{Z-Y}\}, 2Sp\{\rho(t)S_{Z}^{Z-Y}\}\} \),

i.e. C-numbers. Our equation \( \dot{\vec{S}}^{i-j} = \left[ \vec{a} \times \vec{S}^{i-j} \right] \) is of the operator type in contrast to the approaches of \([10],[21]\), and it is the vector operator \( \vec{S}^{i-j}(t) \), which contains the time dependence determined by the solutions of (7), rather than the density matrix \( \rho \).

The problems of the pulsed EPR can be solved more comfortably in the frame rotating with the frequency of the varying field around the \(Z\) axis (RF). Having in mind the subsequent transition to RF, the full UT STS Hamiltonian \( \mathcal{H}^e \) with the account for the varying field directed along the \( K \) axis and acting on the transition \( i-j \), can be rewritten in LF in the form:

\[
\mathcal{H}^* = (E_i - E_j)S_{Z}^{i-j} + (E_k / 2)(\text{matrix}_k) + 4\hbar\omega_{K}^{ij}S_{V}^{i-j} \cos \omega t,
\]

where

\[
(\text{matrix}_x) = \begin{pmatrix}
-1 & 0 & 0 \\
0 & -1 & 0 \\
0 & 0 & 2
\end{pmatrix};
(\text{matrix}_y) = \begin{pmatrix}
2 & 0 & 0 \\
0 & -1 & 0 \\
0 & 0 & -1
\end{pmatrix};
(\text{matrix}_y) = \begin{pmatrix}
-1 & 0 & 0 \\
0 & 2 & 0 \\
0 & 0 & -1
\end{pmatrix},
\]

\(E_i, E_j, E_k\) are the energies of the STS levels. The transition to RF is accomplished by us with the help of the transformation

\[
\mathcal{H}^* = \exp[\mathbf{i}(\pm\omega)S_{Z}^{i-j}/\hbar] \mathcal{H}^e \exp[\mathbf{-i}(\pm\omega)S_{Z}^{i-j}/\hbar]
\]

(the operators in RF are marked off by tildes), and after this transition the spin motion occurs under the action of the Hamiltonian

\[
\mathcal{H}^* = \hbar\omega_0S_{Z}^{i-j} = (E_i - E_j + \hbar\omega)S_{Z}^{i-j} + (E_k / 2)(\text{matrix}_k) + 2\hbar\omega_0^{ij}S_{V}^{i-j},
\]

where the upper and the lower signs are related to the cases \((E_i - E_j) > 0\) and \((E_i - E_j) < 0\), respectively. The equations of motion of STOs with \(\omega_{0ji} = \pm \omega_{0ij}\) in RF have the form:

\[
\begin{align*}
\dot{S}_{X}^{i-j} &= -\left( \pm \omega_0^{ij} \mp \omega \right) S_{Y}^{i-j} + 2\omega_0^{ij}S_{Z}^{i-j} \\
\dot{S}_{Y}^{i-j} &= \pm \left( \omega_0^{ij} \mp \omega \right) S_{X}^{i-j} \\
\dot{S}_{Z}^{i-j} &= -2\omega_0^{ij}S_{V}^{i-j},
\end{align*}
\]

and their solutions are given by the following exact expressions:

\[
\begin{align*}
S_{X}^{i-j}(t) &= S_{X,i}^{i-j}(0) \cos(\omega_0 t) + \\
&+ \left( S_{Y,i}^{i-j}(0) \sin(\theta_{K,i}^{ij}) - S_{Z,i}^{i-j}(0) \cos(\theta_{K,i}^{ij}) \right) \sin(\omega_0 t) \\
S_{Y}^{i-j}(t) &= -S_{X,i}^{i-j}(0) \sin(\theta_{K,i}^{ij}) + S_{Z,i}^{i-j}(0) \cos(\theta_{K,i}^{ij}) \\
&+ S_{X,i}^{i-j}(0) \cos(\theta_{K,i}^{ij}) \sin(\omega_0 t) - \\
&- \left( S_{Z,i}^{i-j}(0) \sin(\theta_{K,i}^{ij}) - S_{Y,i}^{i-j}(0) \cos(\theta_{K,i}^{ij}) \right) \cos(\omega_0 t) \\
S_{Z}^{i-j}(t) &= S_{X,i}^{i-j}(0) \sin(\theta_{K,i}^{ij}) + S_{Y,i}^{i-j}(0) \cos(\theta_{K,i}^{ij}) \\
&+ S_{Y,i}^{i-j}(0) \sin(\theta_{K,i}^{ij}) \sin(\omega_0 t) + \\
&+ \left( S_{Z,i}^{i-j}(0) \sin(\theta_{K,i}^{ij}) - S_{X,i}^{i-j}(0) \cos(\theta_{K,i}^{ij}) \right) \sin(\omega_0 t),
\end{align*}
\]

where \(\omega_{0ji} = \sqrt{[\pm \omega_0^{ij} \mp \omega]_+^2 + [2\omega_0^{ij}]^2}\):

\[
\cos(\theta_{K,i}^{ij}) = \pm \left( \omega_0^{ij} \mp \omega \right) / \omega_{0ji},
\]

\[
\sin(\theta_{K,i}^{ij}) = 2\omega_0^{ij} / \omega_{0ji},
\]

and \(S_{X,i}^{i-j}(0), S_{Y,i}^{i-j}(0), S_{Z,i}^{i-j}(0)\) are the values of \(S_{X,Y,Z}(t)\) at the initial moment.

We would like to note that the exact expressions (11), (12), which are valid for the STS in ZF, are the analogues of the solutions of the Bloch equations without decay in the corresponding RF for the usual Zeeman spin system in the nonzero constant magnetic field. The difference is that the operator \(S_{V}^{i-j}(t)\) corresponds to the Bloch variable \(u\) and the operator \(S_{X}^{i-j}(t)\) corresponds to the Bloch variable \(v\) – for the comparison see (III.16) in [12].

IV. Exact Calculation of FID Signal and Approximate Calculation of Two-Pulse Spin Echo Signal from STS in ZF

The calculation of the free induction decay (FID) signal from the photoexcited STS after the action of a single pulse.
of the microwave (MW) field with \( t_p \) duration, polarized along the \( \mathbf{K} \) axis, under conditions of the arbitrary distribution of spin isochromats of \( ij \) transition can be an example of the use of (11), (12). The turning on of the MW pulse leads to the transition to RF, where STOs look like
\[
\begin{align*}
\tilde{S}^{ij}_x(t') &= S^{ij}_x \cos (\pm \omega t') - S^{ij}_y \sin (\pm \omega t') \\
\tilde{S}^{ij}_y(t') &= S^{ij}_x \sin (\pm \omega t') + S^{ij}_y \cos (\pm \omega t') \\
\tilde{S}^{ij}_z(t') &= S^{ij}_z.
\end{align*}
\] (13)

The measured voltage \( U_{\text{ave}}^{ij} \) on the receiver/transmitter coil, oriented along the \( \mathbf{K} \) axis, at the moment \( t' \), counted out from the end of the pulse, is proportional to \( \left\{ M_K(t') \right\} \), where the average value \( \left\langle M_K(t') \right\rangle \) should be calculated as \( \left\langle M_K(t') \right\rangle = (-2n g_K \mu_B) S \rho S^{ij}_y(t') \).

Here, \( \left\langle M_K(t') \right\rangle \) includes the component \( S^{ij}_y(t') \) of the vector \( \tilde{S}^{ij}_y(t') \) in LF, which is equal to
\[
\begin{align*}
\tilde{S}^{ij}_x(t') &= -S^{ij}_x \sin (\pm \omega t') + \tilde{S}^{ij}_y(t') \cos (\pm \omega t'),
\end{align*}
\]
where \( \tilde{S}^{ij}_x(t') \) are determined by the free precession values
\[
\begin{align*}
\tilde{S}^{ij}_x(t') &= S^{ij}_x(t' = 0) \cos \left[ \left( \pm |\omega_0^{ij}|-\omega \right) t' \right] \\
-\tilde{S}^{ij}_y(t' = 0) \sin \left[ \left( \pm |\omega_0^{ij}|+\omega \right) t' \right],
\end{align*}
\] (14)

with \( \tilde{S}^{ij}_x(t' = 0) = \tilde{S}^{ij}_x(t_p) \). During the calculation of \( \tilde{S}^{ij}_x(t_p) \), it should be taken into account that the following correlations take place at the moment of the switching on of the MW pulse: \( \tilde{S}^{ij}_x(0) = S^{ij}_x \), and at the taking of the trace in \( \left\langle M_K(t') \right\rangle \) it should be considered that
\[
\begin{align*}
\left\langle S^{ij}_x \right\rangle &= S \rho \left\langle S^{ij}_y \right\rangle = (1/2) (P_i - P_j), \\
\left\langle S^{ij}_y \right\rangle &= S \rho \left\langle S^{ij}_y \right\rangle = 0.
\end{align*}
\]
Therefore, at the calculation of \( \tilde{S}^{ij}_x(t_p) \) only the terms with \( S^{ij}_y \) should be allowed for.

As a result,
\[
\begin{align*}
\tilde{S}^{ij}_x(t_p) &= S^{ij}_x \sin \theta^{ij}_K \sin \omega^{ij}_{\text{eff}} t_p, \\
\tilde{S}^{ij}_y(t_p) &= S^{ij}_x \sin \omega^{ij}_{\text{eff}} \sin \left( \omega^{ij}_{\text{eff}} t_p / 2 \right), \\
\tilde{S}^{ij}_z(t_p) &= S^{ij}_x - 2S^{ij}_y \sin^2 \theta^{ij}_K \sin^2 \left( \omega^{ij}_{\text{eff}} t_p / 2 \right).
\end{align*}
\] (15)

Further, we suppose that the inhomogeneous broadening of the EPR lines (the variations of frequencies of their isochromats) of the STS transitions is the nature of the free induction decay, observable on STS transitions. At that, we use the arbitrary distribution \( g(\Delta \omega^{ij}) \) of the frequencies of spin isochromats, where \( \Delta \omega^{ij} \) is the difference between the isochromate frequency and the central frequency \( \omega^{ij}_0 \) of the i-j transition. Carrying out the described calculations and producing then the averaging of the result with the arbitrary distribution of spin isochromats \( g(\Delta \omega^{ij}) \), we obtain that the FID voltage is proportional to the value
\[
U_{\text{ave}}^{ij}(t) \propto \left( -\left\langle M_K(t) \right\rangle \right) \left( -\left\langle M_K(t) \right\rangle \right) = \pm n g_K \mu_B (P_i - P_j) \left| \sin \left( \pm |\omega^{ij}_0| \right) \right| f_{X\text{ean}}(t) - f_{Y\text{ean}}(t) - \sin \left( \pm |\omega^{ij}_0| \right) \left[ f_{X\text{ean}}(t) + f_{Y\text{ean}}(t) \right],
\] (16)

where
\[
\begin{align*}
&f_{X\text{ean}}(t) = \int_{-\infty}^{\infty} \sin \theta^{ij}_K \sin (\omega^{ij}_{\text{eff}} t) \\
&\times \cos \left( \Delta \omega^{ij} t \right) g(\Delta \omega^{ij}) d(\Delta \omega^{ij}); \\
&f_{Y\text{ean}}(t) = \int_{-\infty}^{\infty} \sin \omega^{ij}_{\text{eff}} \sin^2 (\omega^{ij}_{\text{eff}} t_p / 2) \\
&\times \cos \left( \Delta \omega^{ij} t \right) g(\Delta \omega^{ij}) d(\Delta \omega^{ij});
\end{align*}
\] (17)

\[
\omega^{ij}_{\text{eff}} = \sqrt{\left( \pm |\omega^{ij}_0| \right)^2 + 2\omega^{ij}_k^2}.
\] (18)

By the same procedures, the exact signal of the two-pulse echo signal from STSs in ZF can be obtained. However, (16)-(18) have much more simple form in the case of the full excitement of one of the STS transitions. Then during the MW pulses one has \( \cos \theta^{ij}_K \approx 0; \sin \theta^{ij}_K \approx 1 \). In addition, now we suppose that the frequency distribution of the isochromates is described by a Gaussian with the width \( \Delta^{ij} \). As a result, the time dependence of \( \left\langle M_K(t) \right\rangle \) during FID has the form of the following linear oscillation:
\[
\begin{align*}
&\left\langle M_K(t) \right\rangle_G = (-n g_K \mu_B) (P_i - P_j) \sin \left( \theta_{kD} \right) \\
&\times \sin \left( \pm |\omega^{ij}_0| \pm \omega + \Delta \omega^{ij} t \right) \exp \left[ -t^2 \left( \Delta^{ij} / 2 \right)^2 \right],
\end{align*}
\] (19)

where \( \theta_{kD} = 2\omega^{ij}_k t_p \); the index G denotes the averaging over the Gaussian distribution of isochromates. The corresponding FID signal \( U_{\text{ave}}^{ij}(t) \) can be easily obtained from (19). If the MW field has \( \delta^{ij} \)
detuning from $|\alpha_i^{v-1}|$ i.e. $\omega = |\alpha_i^{v-1}| + \delta_i^{v-1}$, then
\[
\sin\left(\pm |\alpha_i^{v-1}| \mp 2\omega t\right) = \mp \sin\left(|\alpha_i^{v-1}| + 2\delta_i^{v-1} + t\right)
\]
with the upper sign valid for $\alpha_0^{X-x}, \alpha_0^{Y-Z}$, and the lower sign valid for $\alpha_0^{Z-X}$. In the case of the exact tuning ($\delta_i^{v-1} = 0$) (19) without our additional coefficient $\left(-n\gamma_i^{v-1} \mu_B\right)$ coincides with (11)-(13) from [6]. The difference is that (19) takes into account the time dependence of the FID signal, caused by the distribution of the frequencies of the i-j transition, and the possibility of the g-factor anisotropy along the different molecular axes. However, the initial (19) allows for the MW field detuning. Note that at the photoexcitation of the organic molecules the probabilities $P_x, P_y, P_z$ can differ strongly from their equilibrium values, determined by the Boltzmann law. They reflect the spin polarization reached because of the photoexcitation. For instance, for STSs of pentacene molecules in p-terphenyl, after the laser flash, which in [7], [8] precedes the MW pulse, the correlation $P_x : P_y : P_z = 0.76 : 0.16 : 0.08$ [7] takes place. The approximation of the full excitation is further used by us for the calculation of the two-pulse echo (with the pulse separation $\tau$) signal from STS. At the neglecting the pulse durations as compared to the value of $\tau$, the calculation of the time dependence of $\tilde{S}_Y^{v-1}(t)$ after the second pulse results in the expression:
\[
\tilde{S}_Y^{v-1}(t) = S_L^{v-1} \cos \theta_{k_1} \sin \theta_{k_2} \\
\cdot \sin\left(\pm |\alpha_i^{v-1}| \mp \omega t\right) - \\
\cdot S_L^{v-1} \sin \theta_{k_1} \sin^2 \left(\theta_{k_2} / 2\right) \\
\cdot \cos\left(\pm |\alpha_i^{v-1}| \mp \omega (t - \tau)\right),
\]
where $t$ is counted out from the end of the second MW pulse: $\theta_{k_1,k_2} = g_i^{v-1} \mu_B h^{-1} 2B_1^{v-1} t_{p_1,p_2}$; $t_{p_1,p_2}$ are the durations of the first and the second pulses, respectively. The first term of (20) conditions the second FID, and the second term conditions the two-pulse echo signal. Passing over to LF and carrying out the averaging of $\tilde{M}_k(t)$ with the Gaussian distribution of the isochromate frequencies of the width $\Delta^{v-1}$, with accounting only for the second term of (20), we obtain that the spin echo signal is proportional to the following value:
\[
U_{\text{ind}}^{v-1}(t)_{\text{echo}} \propto \left\langle -\tilde{M}_k(t) \right\rangle_g = \\
= -n g_i^{v-1} \mu_B (P_i - P) \sin \left(\theta_{k_2} \right) \sin^2 \left(\theta_{k_2} / 2\right) \\
\cdot \left(\pm |\alpha_i^{v-1}| \mp 2\omega \right) \cos\left(\pm |\alpha_i^{v-1}| \mp 2\omega (t - \tau)\right) \\
\cdot \exp\left(\pm |\alpha_i^{v-1}| \mp 2\omega (t - \tau)^2 / 2\right).
\]
Note that the dependence on the angles $\theta_{k_1}, \theta_{k_2}$ in (21) repeats the similar dependence in the Zeeman spin system in a constant field (see (1.15) of [22]), but with the difference that the angle values are doubled and the magnetization accomplishes linear oscillations. This fact was mentioned also in [7] concerning the FID signal from STS. The dependence of the type $\sin \left(\theta_{k_1}\right) \sin^2 \left(\theta_{k_2} / 2\right)$ on the amplitude of the linearly polarized MW field and pulse durations is confirmed by the results of the experiments of the paper [9]. There, the maximum of the two-pulse echo signal at the identical MW pulses was observed at $\theta_{k_1} = \theta_{k_2} = 2\pi / 3$, as it was in the case of Zeeman spin system in a constant magnetic field.

V. FID SIGNAL AT THE ABSENCE OF NUTATION AND STEADY-STATE DYNAMIC SUSCEPTIBILITIES AT THE ABSENCE OF SATURATION FROM STS IN ZF

Equation (19) is valid in the case of a sufficiently strong MW pulse, which is able both to cause the nutation of the single transition spin vector and to provide the full excitation of the STS transition. Then its detuning with respect to the frequency of one of the STS transitions plays no role for the FID amplitude. However, there is a possibility to observe the change of the FID amplitude at the gradual approach of the pulse frequency $\omega$ to $|\alpha_i^{v-1}|$ at the action on STS of a sufficiently weak and short MW pulse (which is not able to cause the nutation of the single transition spin vector). It should be mentioned that this possibility realizes under conditions $g_i^{v-1} \mu_B h^{-1} 2B_1^{v-1} t_p < 1$ nonoptimal for the observation of FID. Under such conditions, one can take $\tilde{S}_Z^{v-1} = 0$ in equations (7). Then the equations of the forced motion of the components of the sample magnetization, brought in [17], have the improved form:
\[
\left\langle \tilde{M}_k(t) \right\rangle + \left(\alpha_i^{v-1}\right)^2 \left\langle M_k(t) \right\rangle = \\
\mp n \left(g_i^{v-1}\right)^2 \mu_i^{v-1} \left(P_i - P\right) 4B_1^{v-1} \cos \omega t.
\]
Equations (22) describe the forced swinging of linear oscillators without decay – the case, considered in [23, pp. 13-15]. With the help of the corresponding formula from [23], the result of the pulse action and further – the FID signal (the improved version of the result of [17]) can be easily obtained:
\[
U_{\text{ind}}^{v-1}(t)_{\text{FID}} \propto \left\langle -\tilde{M}_k(t) \right\rangle_g = \pm n g_i^{v-1} \mu_B (P_i - P) \\
\cdot \left(g_i^{v-1} \mu_B h^{-1} 2B_1^{v-1} t_p \alpha_i^{v-1} \right) \cos\left(\pm |\alpha_i^{v-1}| t\right) \\
\cdot \exp\left(-t^2 \left(\Delta^{v-1}\right)^2 / 2\right).
\]
The periodic dependencies of the FID amplitude value both on the detuning, and on the exciting pulse duration are the interesting consequences of (23).

We can also describe the steady-state evolution of the magnetization components of a sample with STS in ZF under the action of a weak (nonsaturating) varying field, directed along the axis $K = X, Y, Z$ using the approximate equations (22), the l.h.s. of which are phenomenologically supplemented with the relaxation terms $2\langle M_k(t)\rangle/T_{2K}$ (see [24]). Such suggestion implies that $T_{2K}$ coincides with the phase memory time $T_m$ of STS spins, taking part in i-j transition [10]; at that $T_{2K}$ are different for the different transitions [24]. It is seen from the obtained equations that the linear steady-state oscillations of the $K$-th component of the full magnetization of a sample $\langle M_K(t)\rangle$ are caused by the varying with the frequency $\omega \approx |\alpha_0^+/\rangle$ magnetic field polarized along the $K$ axis, where $K, i, j = X, Y, Z = Y, Z, X = Z, X, Y$. The solution of these equations allows writing the complex tensor of the steady-state dynamic susceptibility of STS to a varying field:

$$
\chi = \begin{pmatrix}
X_{XX} & 0 & 0 \\
0 & X_{YY} & 0 \\
0 & 0 & X_{ZZ}
\end{pmatrix},
$$

(24)

where

$$
X_{KK} \equiv X_{KK}^* - iX_{KK}^* = \mu_0 \left[ -n \frac{\left(g_K^\prime\right)^2}{\hbar^2} \right] \left( P_i - P_j \right) \frac{\left( |\alpha_0^+/\rangle - \omega - i(T_{2K})^{-1} \right)^2 + (T_{2K})^2}{\left( |\alpha_0^+/\rangle - \omega \right)^2 + (T_{2K})^2},
$$

(25)

the signs $\pm$ are related to $|\alpha_0^+/\rangle = \pm |\alpha_0^-\rangle$. The values of $X_{KK}^* \cdot X_{KK}^*$ can be registered with the help of the receiver/transmitter coil along the $K$ axis. Equation (25) is the improved version of the result of [17].

VI. CONCLUSION

Summarizing, the following results are obtained in the given paper:

The unitary transformation operator is obtained, which diagonalizes the axially asymmetric Hamiltonian ($E \neq 0$) conditioning the zero field spitting (ZFS) of the spin triplet state (STS) levels in the basis of the usual triplet wave functions. This enabled us to use the single transition spin operators (STOs) for the formulation of this Hamiltonian and that of the spin interaction with the varying magnetic field. The equations of motion for the components $S_{ij}^{-}(t), S_{ij}^{+}(t), S_{ij}^{0}(t)$ of the STO vector $\mathbf{S}^{-}(t)$ are derived for the STS subjected to the action of these two Hamiltonians. The obtained equations written in terms of one averaged equation for STO vector $\mathbf{S}^{-}(t)$ appeared to be a particular case (for STS) of the universal equation from [21]. As well as that, our equation for $\mathbf{S}^{-}(t)$ is analogous to the Bloch equation without decay for the usual Zeeman system in a constant magnetic field and a transverse to it varying field. However, our equation is of the operator type and is obtained with the help of the quite different method than that in [21]. All kinds of spin "acrobatics" (the term from [6]), which was ever described for Zeeman spin system using usual Bloch equations, can be described for only one excited STS transition using the equations of motion derived here, if the population probabilities of the corresponding levels are not artificially equalized. Hence, the method of STS treatment in a convenient and evident presentation is elaborated in the given paper for axially asymmetric ZFS with a few demonstrative examples – to the best of our knowledge, not described analytically before (except for the FID signal in the case of nutation and of the full excitement of the corresponding transition). It should be also noticed that the motion of the full sample magnetization, investigated with the help of the solutions of the STO equations, appeared to have the quite different character than that of the motion of the STO vector $\mathbf{S}^{-0}(t)$. Namely, the motion of magnetization has the linear character, as it was mentioned in [18]. Here, in terms of this magnetization the signals of FID (as in the case of nutation, so without it), of the two-pulse spin echo and of the values of the dynamic susceptibilities to a weak varying field are calculated for STS in ZF. The abovementioned results can be applied in ZF to any physical object mentioned in Introduction, including nuclear quadrupole resonance for nuclei with spin 1 (especially, $^{14}$N) and the nonzero asymmetry parameter, what is important for the monitoring of the nitrogen-containing explosives and drugs [4].

APPENDIX

The demonstration of the full coincidence of the result of our approach at the derivation of the particular form (for STS) of the equation $\hat{\mathbf{r}} = [\hat{a} \times \hat{F}]$ of [21] with the results obtained in [10] and for the demonstration of the definite advantage of our approach is the aim of this Appendix.

Let us switch from the approach of [10] to that of ours. This transition is realized by means of the transition to the unitary transformed operators: $\mathbf{S}_{X,Y,Z} \rightarrow \mathbf{S}_{X,Y,Z}$. At that, it is necessary to use the usual triplet eigenfunctions $|T_{1}\rangle, |T_{0}\rangle, |T_{1}\rangle$ instead of these of [10]. The last transition, accounting for the different sequence of the energy levels in [10], where the $X$ and $Z$ axes and $X$ and $Z$ levels traded their places compared to our axes and levels, takes place in the following manner: $|T_{Z}\rangle \rightarrow |T_{1}\rangle; |T_{Y}\rangle \rightarrow |T_{0}\rangle$. In such a case, the unitary operator for the diagonalization of the ZFS Hamiltonian and the UT ZFS Hamiltonian itself are presented by the following matrices

\[ \begin{pmatrix}
X_{XX} & 0 & 0 \\
0 & X_{YY} & 0 \\
0 & 0 & X_{ZZ}
\end{pmatrix}. \]
An investigation using formulae (28) of Chapter 2 of [10] with the corrected expression there. Here, the vector \( \hat{r} \) can be defined as

\[
\hat{r} = \frac{\vec{\partial} \times \vec{F}}{|\vec{\partial} \times \vec{F}|}
\]

of [10] after the described transition and the vector \( \vec{\partial} \) has the components \( (0, 4\alpha_0^{X,Y} \cos \omega t, \alpha_0^{Z,Y}) \) in our notations, what is seen using (5) from [21], where now \( V = g_X \mu_B 2B_\text{ix} \cos \omega t S_X^{i,i,X} \).

The above formulæ for \( F \) should be compared with the formulæ (28)-(30) of Chapter 2 of [10] with the corrected misprint in \( r_i \) expression there. The corresponding equation \( \hat{r} = \frac{\vec{\partial} \times \vec{F}}{|\vec{\partial} \times \vec{F}|} \) of [10] should be compared with (7) of the given paper, written for \( i - j = Y - Z \) and with the accounting for the facts that \( S_{X,Y,Z}^{i,j} = S_{i,j,X} \); \( S_{i,j,Y}^{i,j} = -S_{Y,Z}^{i,j} \); \( \alpha_0^{X,Y} = -\alpha_0^{Y,X} \); \( \alpha_0^{Z,Y} = -\alpha_0^{Y,Z} \). Then the full coincidence is seen with the difference that, in our opinion, it is much more comfortable to handle with the STOs when describing the complicated pulse EPR experiments.

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