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General Inverse Problem Solution for Two-Level Systems and Its Application to Charge Transfer

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Abstract: Two-level quantum systems are building blocks of quantum technologies, where the qubit is the basic unit of quantum information. The ability to design driving fields that produce prespecified evolutions of relevant physical observables is crucial to the development of such technologies. Using vector algebra and recently developed strategies for generating solvable two-level Hamiltonians, we construct the general solution to the inverse problem for a spin in a time-dependent magnetic field and its extension to any two-level system associated with fictitious spin and field. We provide a general expression for the field that drives the dynamics of the system so as to realize prescribed time evolutions of the expectation values of the Pauli operators and the autocorrelation of the Pauli vector. The analysis is applied to two-state charge transfer systems, showing that the charge transfer process can be seen as a motion of the state of the associated fictitious qubit on the Bloch sphere, and that the expectation values of the related Pauli operators describe the interference between the two differently localized electronic states and their population difference. Our formulation is proposed as a basic step towards potential uses of charge transfer in quantum computing and quantum information transfer.

Keywords: inverse problem; spin in time-dependent magnetic field; two-level model; charge transfer; electronic coupling; Bloch sphere

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1. Introduction

Realizations of coherent quantum dynamics are the subject of ever-growing interest in fields of investigation ranging from charge and excitation dynamics in biochemical and biophysical systems [1–12] to carbon (nano)materials [13–15], nanotechnology [11,16–18], and novel quantum technologies [18–22]. In all these research fields, the most basic building block of systems that involve (at least partial) coherent quantum dynamics is the qubit, and the control of its dynamics by appropriate driving fields plays a central role in new quantum technologies [22–24], including quantum information processing (where charge, spin, or more generally quantum state transfer is usually involved) [10,11,21,24–31], quantum computing [22,23,32–36], quantum metrology [21,29,37], and sensing [21,38–41].

Apart from the spin qubit, which consists of a spin in a magnetic field, a charge qubit can be realized, e.g., by the two charging states of a superconducting island characterized by the absence/presence of an excess Cooper pair [42], or by two electronic wave functions of a quantum dot (which can also correspond to different charging states of the quantum dot) [43], or by a molecule in an optical microcavity [17], or still by a charge-transfer (CT) system [8]. Rapid technological advances are increasingly enabling the construction, robustness, control, and measurability of qubits [14,17,43].

The use of quantum two-level systems [14,22,44] requires their precise initialization and measurement [20], as well as the ability to control their time evolution (especially at avoided crossings in the case of spin, charge, excitation, and information transfer) [8,19,45–52]. Individual spins in generally variable magnetic fields and CT systems that satisfy the two-state approximation offer many relevant opportunities for the implementation of two-level systems and the observation of coherent quantum dynamics: the former thanks to their generally weak coupling to the environment, which can also allow the persistence of quantum coherence at room temperature [15]; the latter due to their ubiquity in biochemical and biophysical systems, and the possibility of using avoided crossing to control their dynamics [1,2,7,8,47,52–54]. Furthermore, as long recognized [55], any two-level system that does not involve a ½-spin particle can still be described as a fictitious spin in a fictitious field, which allows the application to CT [8] of methods initially (implicitly or explicitly) conceived for studying ½ spins in time-dependent magnetic fields [56–61].

The above considerations stress the relevance of theoretical models in which the two-level system driven by a generally variable field or interaction is described by a time-dependent Hamiltonian. The field can be externally applied or be expression of the interaction of the given qubit with other qubits or other components of the surrounding environment [45,49,62–69]. Time-dependent parameters determine the evolution of two-level or multi-level systems related to CT [8,70], spin population transfer and dynamics in general [60,71,72], quantum computing [23,34,36,73], quantum information processing [74–78], NMR spectroscopy [79], quantum plasmonics [80], and so forth.

The study of (two-level) systems described by time-dependent Hamiltonians is relevant in terms of both direct and inverse problems [81]. In the first case, the aim is to find the exact time evolution of a system described by a known Hamiltonian (e.g., a spin subject to an external field with known time dependence). This has stimulated the search for conditions under which a time-dependent Hamiltonian problem can be exactly solved analytically [82]. In the second case, the goal is finding the time-dependent field that produces a desired unitary transformation of the system, i.e., a prespecified dynamic evolution. For example, this is a typical problem in quantum computing, where quantum gates are used to evolve qubits to final objective states and coherent control of the qubit evolution is crucial to implement reliable and reproducible computations. The quantum computing machinery must work whatever the input, producing the appropriate unitary transformation of any given initial set of qubit states [34,83]. In these contexts, apart from the central role of qubits as units of information, one-qubit models are often implemented; for example, they are exploited in Ref. [84], where the time evolution of spin chains periodically driven by light is simulated on quantum computers.

In a more general context, which also includes machine learning techniques (e.g., used to learn quantum states [85,86]), expectation values of physical observables and possibly their correlations are obtained from experiments and represent the starting point of an inverse problem. In the specific case of quantum state learning, the problem is to estimate an unknown quantum system, starting from measurements of observables on the system [85], but in many other situations the accent is on the evolution of the system, i.e., on how to obtain the desired dynamic evolutions of the measured quantities by suitably engineering the system, namely, the Hamiltonian that represents the system (in particular, dynamical invariants can also be used for the engineering purpose [87]). When the reverse engineering concerns spins in variable magnetic fields, the expectation values of the Pauli matrices, or the corresponding spin operators, play the most significant role in describing the dynamics [88]. Their importance holds for other types of two-level systems, since they can be formulated in terms of a fictitious spin, as is shown below.

In this study, we entirely solve the inverse problem concerning two-level systems for an input that consists of the time-dependent expectation values of the Pauli operators and one of their possible correlations. First, through a convenient parameterization of the time evolution operator, we formulate the expression for the driving magnetic field that generates a prescribed unitary evolution (Section 2). After that, we solve the inverse problem of finding the time-dependent field that produces any prescribed evolution of the expectation values of the Pauli operators and the time autocorrelation of the Pauli vector (its real part is sufficient indeed). The analysis also affords the expressions for the matrix elements of the evolution operator in terms of such quantities, thus also completely solving the dynamical problem for the engineered Hamiltonian (Section 3). The mapping of a generic two-level system to a fictitious spin in a fictitious magnetic field is used in Section 4 to highlight the applicability of the approach to any two-level system, such as the fundamental two-state CT system investigated in Section 5. The analysis (i) clarifies the meaning and significance of the chosen observable expectation values in the case of CT processes and (ii) transparently interprets the CT as the motion of a point representing a qubit state on the Bloch sphere, with relevance to potential quantum information and quantum computing uses of CT processes, and opportunities for generalizations briefly discussed in the conclusive Section 6.

2. Spin in a Variable Magnetic Field: Solving the Inverse Problem

Using Pauli matrices $\boldsymbol{\sigma} = (\sigma_1, \sigma_2, \sigma_3)$, the Hamiltonian $H(t)$ of a spin-1/2 particle in an arbitrary time-dependent magnetic field $\mathbf{B}(t)$ is given by:

$$\frac{H(t)}{\hbar} = \mathbf{B}(t) \cdot \boldsymbol{\sigma} = \begin{pmatrix} B_3 & B_1 - iB_2 \\ B_1 + iB_2 & -B_3 \end{pmatrix}, \tag{1}$$

where the simplifying notation $\mathbf{B}(t) = -\frac{2}{\gamma}\mathbf{B}(t)$ is used, γ is the gyromagnetic ratio of the system, and $\hbar$ is the reduced Planck constant. More generally, and also in our application below, this Hamiltonian model represents a fictitious spin in a fictitious magnetic field [8,55].

The time-evolution operator for $H(t)$ belongs to SU(2) and, e.g. exploiting Cayley–Klein parameters, its common parameterization reads [59,89,90].

$$U(t) \equiv U(t,0) = \begin{pmatrix} a(t) & b(t) \\ -b*(t) & a*(t) \end{pmatrix}, \tag{2}$$

where $|a(t)|^2 + |b(t)|^2 = 1$ and, since $U(0) = 1$ (i.e., the identity operator, or its matrix representation), it is $a(0) = 1$ and $b(0) = 0$.

Here, we introduce the following further parameterization:

$$U(t) = u_0(t)1 - i\,\mathbf{u}(t) \cdot \boldsymbol{\sigma}. \tag{3}$$

In Equation (3), $(u_0, \mathbf{u})$ are four real functions of time (where there is no ambiguity, the time dependence of the quantities is not explicitly shown to simplify the notation) related to a and b by the relations.

$$a = u_0 - i\,u_3, \quad b = -i\,(u_1 - i\,u_2) = -u_2 - i\,u_1. \tag{4}$$

Furthermore, the unitarity of U imposes the constraint:

$$u_0^2 + u^2 = 1 \tag{5}$$

on the norm of $(u_0, \mathbf{u})$, with u denoting the modulus of $\mathbf{u}$.

The field $\mathbf{B}$ is related to $(u_0, \mathbf{u})$ by (see Appendix A).

$$u'_0 = -\mathbf{B} \cdot \mathbf{u}, \quad \mathbf{u}' = u_0\mathbf{B} + \mathbf{B} \times \mathbf{u}, \tag{6}$$

where Lagrange notation is used for derivatives. From Equations (5) and (6), we derive the following expression for $\mathbf{B}$ in terms of $(u_0, \mathbf{u})$ (Appendix A):

$$\mathbf{B} = u_0\mathbf{u}' - u'_0\mathbf{u} + \mathbf{u} \times \mathbf{u}'. \tag{7}$$

Equation (7) solves the inverse problem of finding a time-dependent field that generates a prescribed unitary evolution in the case of a single spin, which is the prototypical qubit, with relevance to fields of investigation ranging from control theory [23,46,83,91,92] to quantum computing, especially where the implementation of quantum gates is involved [20,32,46,83,93]. However, in Section 3 we will solve the inverse problem in the most practicable experimental way, that is, starting from a desired evolution of the expectation values of observables amenable to measurement.

3. Finding (Engineering) the Field That Produces the Desired Observable Evolution

Since the system under study is a (pseudo)spin, Pauli matrices represent the fundamental observables of interest. In this section, we determine the time-dependent magnetic field that makes their expectation values and correlations evolve in a prefixed way.

The expectation values of Pauli matrices σ_1, σ_2, and σ_3 are compactly written as:

$$\mathbf{v}_t \equiv \mathbf{v}(t) \equiv \langle \boldsymbol{\sigma} \rangle(t) = \mathrm{Tr}\Big[\boldsymbol{\sigma} U(t) \rho_0 U^\dagger(t)\Big], \tag{8}$$

where $\rho_0 \equiv \rho(0)$ is the density matrix that describes the initial state of the qubit. In particular,

$$\mathbf{v}_0 \equiv \mathbf{v}(0) = \mathrm{Tr}[\boldsymbol{\sigma}\rho_0]. \tag{9}$$

$\mathbf{v}_t$ depends on $\mathbf{v}_0$ and the time evolution operator. Using Equations (3) and (8), we obtain (Appendix B).

$$\mathbf{v}_t = \mathbf{v}_0 + 2u_0\, \mathbf{u} \times \mathbf{v}_0 + 2\mathbf{u} \times (\mathbf{u} \times \mathbf{v}_0). \tag{10}$$

It is worth noting that scalar multiplication of Equation (10) by $\mathbf{u}$ readily gives:

$$\mathbf{u} \cdot \mathbf{v}_t = \mathbf{u} \cdot \mathbf{v}_0, \tag{11}$$

i.e., the displacement of $\mathbf{v}_t$ is always orthogonal to $\mathbf{u}$. Also, as expected (Appendix B),

$$v_t^2 = v_0^2. \tag{12}$$

Part of the dependence of $\mathbf{B}$ on $\mathbf{v}_t$ results from the relationship between the expectation values of the Pauli operators and the components of $\mathbf{B}$. This relation leads to:

$$\mathbf{v}'_t = 2\mathbf{B} \times \mathbf{v}_t, \tag{13}$$

which is the analogue of the macroscopic Bloch equations in our single spin model. Vector multiplication of Equation (13) by $\mathbf{v}_t$ on the right, together with the use of Equations (A6) and (12), gives:

$$\mathbf{B} = \frac{1}{2v_0^2}\Big[\mathbf{v}_t \times \mathbf{v}'_t + 2(\mathbf{B} \cdot \mathbf{v}_t)\mathbf{v}_t\Big]. \tag{14}$$

Equation (13) shows that the component of the variable magnetic field $\mathbf{B}$ parallel to $\mathbf{v}_t$ does not influence the first time derivative of $\mathbf{v}_t$. However, this component must also be constrained to achieve a specific time evolution of the system, as described, for example, by a given $U(t)$ relevant for quantum computing tasks or as required to produce desired correlations of some (at least one: vide infra) physical quantities. From Equation (13) it is easily realized that for a time-dependent field the component of $\mathbf{B}$ parallel to $\mathbf{v}_t$ (which is named $\mathbf{B}_\parallel$ in the following) influences the evolution of $\mathbf{v}_t$. For example, the derivation of Equation (13) with respect to t, the subsequent substitution of the expression (13) for $\mathbf{v}'_t$, and the use of Equation (A6) give $\mathbf{v}''_t = 2\mathbf{B}' \times \mathbf{v}_t + 4B_\parallel v_t \mathbf{B} - 4B^2\mathbf{v}_t$, with evident contributions from $\mathbf{B}_\parallel$. Note that also the first term in this expression is generally nonzero. This is understood considering that, since $\mathbf{B}_\parallel(t)$ is parallel to $\mathbf{v}_t$ at any t by definition and $\mathbf{v}'_t \perp \mathbf{v}_t$, $\mathbf{B}'_\parallel(t)$ also has a component which, being parallel to $\mathbf{v}'_t$, is orthogonal to $\mathbf{v}_t$ and therefore contributes to $\mathbf{v}''_t$.

According to Equation (7), the solution of the inverse problem starting from the assignment of $\mathbf{v}_t$ (and in particular of $\mathbf{v}_0$) requires finding the expression of $(u_0, \mathbf{u})$ as a function of $\mathbf{v}_t$ and $\mathbf{v}_0$. However, in general, the relation between $(u_0, \mathbf{u})$ and $\mathbf{v}_t$ cannot be fully disentangled. In Appendix C we show that:

$$\tilde{\mathbf{u}} \equiv \frac{\mathbf{u}}{u_0} = \frac{w(\mathbf{v}_0 + \mathbf{v}_t) + \mathbf{v}_0 \times \mathbf{v}_t}{v_0^2 + \mathbf{v}_0 \cdot \mathbf{v}_t} \tag{15}$$

with

$$w \equiv \tilde{\mathbf{u}} \cdot \mathbf{v}_0 = \pm\sqrt{\frac{v_0^2 + \mathbf{v}_0 \cdot \mathbf{v}_t}{2u_0^2} - v_0^2}. \tag{16}$$

Then, using the equality $\mathbf{u}' = u'_0\tilde{\mathbf{u}} + u_0\tilde{\mathbf{u}}'$ to rewrite Equation (7) as:

$$\mathbf{B} = u_0^2\left(\tilde{\mathbf{u}}' + \tilde{\mathbf{u}} \times \tilde{\mathbf{u}}'\right), \tag{17}$$

after lengthy vector algebra (Appendix C), one obtains:

$$\mathbf{B} = \frac{1}{2v_0^2}\mathbf{v}_t \times \mathbf{v}'_t + \mathbf{B}_\parallel, \tag{18}$$

with

$$\mathbf{B}_\parallel = \left[2u_0^2 w' - \frac{\mathbf{v}_0 \cdot (\mathbf{v}_t \times \mathbf{v}'_t)}{2v_0^2}\right]\frac{\mathbf{v}_t}{v_0^2 + \mathbf{v}_0 \cdot \mathbf{v}_t}. \tag{19}$$

Equations (18) and (19) are consistent with the general expectation from Equation (14) and manifests an incomplete disentanglement of $(u_0, \mathbf{u})$ and $\mathbf{v}_t$ through the presence of u_0. Note that $\mathbf{B}$ depends on the initial condition, since there is only one specific field (and hence Hamiltonian) that produces the desired evolution starting from a given initial spin state.

We can complete our definition of the inverse problem and arrive at a full recipe for finding the required field $\mathbf{B}$ by complementing the initial knowledge of $\mathbf{v}_t$ with that of one correlation of Pauli operators. In the present analysis, we mostly focus on the time autocorrelation of the Pauli vector $\boldsymbol{\sigma}$, which can be written in the form (the derivation is presented in Appendix D).

$$\mathcal{C}(t) \equiv \mathrm{Tr}[\boldsymbol{\sigma}(t) \cdot \boldsymbol{\sigma}\rho_0] = \mathrm{Tr}\left[U^\dagger(t)\boldsymbol{\sigma}U(t) \cdot \boldsymbol{\sigma}\rho_0\right] = 4u_0^2 - 1 + 4iu_0\mathbf{u} \cdot \mathbf{v}_0. \tag{20}$$

Moreover, since observables are usually related to the real or imaginary part of a correlation function, and the strategies for measuring the real parts of correlations of observables are easier to devise than those for their imaginary parts [94,95], we limit ourselves to considering the real part of the correlation function:

$$\mathrm{Re}\mathcal{C}(t) = 4u_0^2(t) - 1. \tag{21}$$

Note that Equation (21) readily follows from Equation (20), since $\mathbf{u}$ and $\mathbf{v}_0$ are both real by construction. Since $U(0) = 1 \Rightarrow u_0(0) = 1$, it is $\mathcal{C}(0) = \mathrm{Tr}\left[\boldsymbol{\sigma}^2\rho_0\right] = 3$ and the acceptable solution for $u_0(t)$ from Equation (21) is:

$$u_0(t) = \frac{1}{2}\sqrt{1 + \mathrm{Re}\mathcal{C}(t)}. \tag{22}$$

Equations (18), (19) and (22) solve the inverse problem of determining the magnetic field $\mathbf{B}$

that produces the desired time evolution of $\mathbf{v}_t$ and Re$\mathcal{C}(t)$ starting from the initial conditions $\mathbf{v}_0$ and $\mathcal{C}(0) = 3$. Furthermore, using Equations (15) and (22) yields (Appendix D).

$$\begin{aligned} \mathbf{u}(t) = &\frac{1}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}\Big\{\tfrac{1}{2}\sqrt{1+\mathrm{Re}\mathcal{C}(t)}\mathbf{v}_0\times\mathbf{v}_t \\ &\pm\sqrt{\tfrac{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}{2}-\tfrac{v_0^2}{4}[1+\mathrm{Re}\mathcal{C}(t)]}(\mathbf{v}_0+\mathbf{v}_t)\Big\} \end{aligned} \quad (23)$$

where the use of the + or − sign depends on the values of the quantities $\mathbf{v}_t$ and Re$\mathcal{C}(t)$, which are preassigned in the inverse problem.

For systems with unitary evolution, inserting (22) and (23) into Equation (3) gives the evolution operator $U(t)$, thus providing the complete description of the dynamics of the spin in the time-dependent magnetic field. Different expressions for $U(t)$ would result from using a different correlation, for example Im$\mathcal{C}(t)$ or any of the fundamental two-observable correlations (Appendix E), whose real parts satisfy the relation.

$$\frac{1}{2}\mathrm{Re}\mathcal{C}_{np}(t) \equiv \mathrm{ReTr}\big[\sigma_n(t)\sigma_p\rho_0\big] = \left(u_0^2-\frac{1}{2}\right)\delta_{np} - u_0\sum_{j=1}^{3}u_j\varepsilon_{jnp} + u_n u_p. \quad (24)$$

4. The Inverse Problem for a General Two-Level System

As pointed out in Section 2, the above analysis can generally be referred to a fictitious spin in a fictitious field **B**, thus being applicable to any two-level system (which can also represent a qubit). Without loss of generality [59], the Hamiltonian matrix can be written as [8,55,59].

$$H(t) = \begin{pmatrix} \Omega(t) & V_{12}(t) \\ V_{12}^*(t) & -\Omega(t) \end{pmatrix} \equiv \begin{pmatrix} \Omega(t) & \frac{1}{2}\omega_0(t)e^{-i\phi(t)} \\ \frac{1}{2}\omega_0(t)e^{i\phi(t)} & -\Omega(t) \end{pmatrix} \quad (25)$$

on the basis of the eigenstates $|1\rangle = \begin{pmatrix}1\\0\end{pmatrix}$ and $|2\rangle = \begin{pmatrix}0\\1\end{pmatrix}$ of the unperturbed (or uncoupled) Hamiltonian ($V_{12} = 0$). Here, $\Omega(t)$ is half the (time-dependent) energy separation between these two states and $V_{12}(t)$ denotes their coupling, which has modulus $\omega_0(t)/2$ and phase $\phi(t)$. One can also use the Pauli matrices and the matrices that represent the ladder operators $\sigma_\pm = \frac{1}{2}(\sigma_1 \pm \sigma_2)$ to recast the Hamiltonian matrix in the two equivalent forms (cf. Refs. [55,96,97]).

$$\begin{aligned} H(t) &= \Omega(t)\sigma_3 + \tfrac{\omega_0(t)}{2}[\cos\phi(t)\,\sigma_1 + \sin\phi(t)\,\sigma_2] \\ &= \Omega(t)\sigma_3 + \tfrac{\omega_0(t)}{2}\left[e^{-i\phi(t)}\sigma_+ + e^{i\phi(t)}\sigma_-\right]. \end{aligned} \quad (26)$$

In units such that $\hbar = 1$, the fictitious magnetic field is related to the matrix elements of the Hamiltonian by the relations [8,55]

$$\begin{cases} B_1(t) = \mathrm{Re}V_{12}(t) = \frac{\omega_0(t)}{2}\cos\phi(t) \\ B_2(t) = \mathrm{Im}V_{12}(t) = \frac{\omega_0(t)}{2}\sin\phi(t) \\ B_3(t) = \Omega(t) \end{cases}, \quad (27)$$

Therefore, the modulus of the field projection onto the xy plane is equal to the modulus of the coupling matrix element, while the field component orthogonal to the xy plane is half the energy difference between the basis states. Furthermore, the phase of the coupling can be derived from the ratio of B_2 and B_1. Equation (27) highlights that the role played by the magnetic field in engineering the dynamics of a spin is played by the energy difference and the coupling between the unperturbed states in the case of a general two-state system. One still needs to find what $\mathbf{v}_t$ and $u_0(t)$ translate into as the inverse problem is reformulated for the general two-level system. This problem is addressed in Section 5, where its solution is further clarified by a formulation for CT systems.

We conclude this section noting that for all cases in which the phase of the coupling satisfies the condition:

$$\phi'(t) = 2\Omega(t) - \mu\omega_0(t) \tag{28}$$

the evolution operator for the system described by the Hamiltonian (25) is already known and has the matrix form [8,59].

$$U(t) = \begin{pmatrix} \sqrt{\frac{\mu^2+\cos^2\Phi(\mu;t)}{1+\mu^2}}\, e^{-i[\frac{\phi(t)}{2}+\vartheta(\mu;t)]} & -i\frac{\sin\Phi(\mu;t)}{\sqrt{1+\mu^2}}e^{-i\frac{\phi(t)}{2}} \\ -i\frac{\sin\Phi(\mu;t)}{\sqrt{1+\mu^2}}e^{i\frac{\phi(t)}{2}} & \sqrt{\frac{\mu^2+\cos^2\Phi(\mu;t)}{1+\mu^2}}\, e^{i[\frac{\phi(t)}{2}+\vartheta(\mu;t)]} \end{pmatrix}, \tag{29}$$

where

$$\Phi(\mu;t) = \sqrt{1+\mu^2}\kappa(t)\,, \quad \kappa(t) = \frac{1}{2}\int_0^t \omega_0(t')dt' \tag{30}$$

and [8]

$$\vartheta(\mu;t) = \begin{cases} \arctan\left[\frac{\mu}{\sqrt{1+\mu^2}}\tan\Phi(\mu;t)\right] & \text{for } t \neq t_{(2n-1)\pi/2}(\mu) \\ (2n-1)\frac{\pi}{2} & \text{for } t = t_{(2n-1)\pi/2}(\mu) \end{cases} \quad (n \in \mathbb{N}). \tag{31}$$

In Equations (29)–(31), $t = t_{\pi/2}(\mu), t_{3\pi/2}(\mu), \ldots$ are the times at which $\Phi(\mu;t) = \frac{\pi}{2}$, $\frac{3\pi}{2}, \ldots$, and μ is a positive real parameter. If $\mu = 0$, Equation (31) is replaced by [8].

$$\vartheta(0;t) = \begin{cases} 0 & \text{for } t_{(-\frac{1}{2}+2n)\pi} \leq t < t_{(\frac{1}{2}+2n)\pi} \\ \pi & \text{for } t_{(\frac{1}{2}+2n)\pi} \leq t < t_{(\frac{3}{2}+2n)\pi} \end{cases} \quad (n \in \mathbb{Z}). \tag{32}$$

The comparison between Equations (3) and (29) provides the expressions for the parameters $(u_0, \mathbf{u})$ in terms of the Hamiltonian parameters. The specific functional forms of $(u_0, \mathbf{u})$ (and therefore of the related Hamiltonian parameters) that correspond to the desired $\mathbf{v}_t$ and $u_0(t)$ are given by Equations (22) and (23). In Section 5, the inverse problem for the two-level system is attacked head-on, after some considerations that then enable a back comparison with the known matrix form (29) of the evolution operator when condition (28) is satisfied.

5. Inverse Charge-Transfer Problem: Controlling the Charge Dynamics

In the framework of CT, $|1\rangle$ and $|2\rangle$ are diabatic electronic states that describe the localization of the excess, transferring charge before and after the CT process. As shown by some of us in a recent study [8], condition (28) can be implemented through a (physical) rotation of the charge donor in a frame where the charge acceptor is fixed, or conversely. After solving the inverse problem proposed hereafter, we will show that it corresponds to physical situations described by $\mu = 0$ in Ref. [8].

Our main purpose in this section is to analyze the meaning and use of the inverse problem solution in the framework of CT, which will also highlight the mapping of CT to the motion of the corresponding qubit on its Bloch sphere in a transparent way. It is worth noting that the states of the system are often denoted $|1\rangle$ and $|2\rangle$ in CT studies and correspond to $|+\rangle = |0\rangle$ and $|-\rangle = |1\rangle$, respectively, in the usual qubit notation.

The Hamiltonian of the CT system has the general form (25). For a given energy difference $2\,\Omega(t)$ between the diabatic electronic states, the modulus of their electronic coupling, $\omega_0(t)/2$, is the determinant of the CT rate when free energy factors are not at play, as we assume here. The electronic state evolution essentially occurs near an avoided crossing, where $2\,\Omega(t)$ becomes small compared to the coupling (Figure 1a). In addition, cases where the charge dynamics involves degenerate localized states are also encountered. For example, CT between two defects in a solid state-like matrix, over suitable temperature ranges, may meet both this condition and the neglect of free energy reorganization. Furthermore, also in this case, the CT dynamics can either be periodic (as in the Rabi oscillation) or not, depending on the time dependence of the electronic coupling between the initial and final states [8].

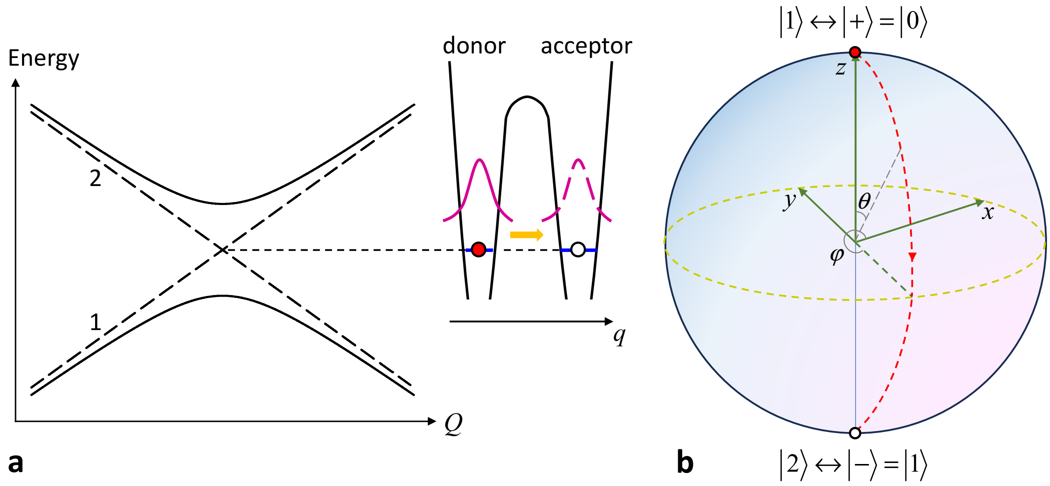

Figure 1. Description of a two-state CT process as a qubit state moving on the Bloch sphere. (**a**) Energy profile of the CT system as a function of a nuclear reaction coordinate or some other time-dependent parameter (such as the amplitude of an applied electric field in a driven CT system [98]) $Q(t)$ describing the system along the CT reaction path [1,8,98]. All quantities in the Hamiltonian matrix (25) depend on time through this parameter; e.g., $\Omega(t) = f(Q(t))$, where f is a suitable function of Q. The dashed lines describe the energies of the diabatic states (that is, the diagonal terms of the Hamiltonian) at varying $Q(t)$ and hence t. The solid lines describe the energies of the adiabatic states, which split near the transition state coordinate, where the diabatic levels cross. Their minimum separation at this coordinate is twice the modulus of the coupling between the diabatic states $|1\rangle$ and $|2\rangle$. Near the transition state, where the charge transition can more easily occur, the diabatic energy difference is negligible compared to the coupling. The inset shows the potential energy profile seen by the transferring charge (e.g., an electron) along its coordinate q, at the transition nuclear coordinate, where the two localized electronic states are degenerate (the corresponding wave functions are schematized in purple). The tunneling probability is determined by the electronic coupling matrix element. (**b**) A coherent CT process can be seen as a motion of the representative point of the system state on a great circle of the Bloch sphere of the fictitious spin associated with the two-level system through Equation (27). If the charge transition probability reaches unity, the corresponding point on the Bloch sphere moves from the north pole to the south pole (red dashed line).

In what follows, we consider degenerate states, and therefore $\Omega(t) = 0$. We also use $\phi = 0$. These parameter choices do not affect our main conclusions, while further simplifying the analysis and maximizing the transparency of the description of a CT process as a motion on the Bloch sphere.

We write the state of the system at a generic time t as

$$|\psi(t)\rangle = C_1(t)|1\rangle + C_2(t)|2\rangle, \tag{33}$$

and the initial conditions are $C_1(0) = 1$, $C_2(0) = 0$, which correspond to a density matrix:

$$\rho_0 = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}. \tag{34}$$

If the charge is finally localized as in state $|2\rangle$, the final state is described by Equation (33) with $C_1 = 0$, $C_2 = 1$. Then, in terms of the associated qubit, the initial and final states are located at the poles of the Bloch sphere (Figure 1b). Inserting the matrix expression (2) for the evolution operator into the matrix equation:

$$\mathbf{C}(t) = U(t)\mathbf{C}(0), \tag{35}$$

where $\mathbf{C}$ is the column vector of C_1 and C_2, for the above initial conditions we obtain:

$$U(t) = \begin{pmatrix} C_1(t) & -C_2^*(t) \\ C_2(t) & C_1^*(t) \end{pmatrix}. \tag{36}$$

Note that $C_1(t)$ and $C_2(t)$ satisfy the normalization condition $|C_1(t)|^2 + |C_2(t)|^2 = 1$ at any t. Furthermore, since a global phase factor of the state vector can be disregarded, $C_1(t)$ is chosen to be real. In general, this may require the inclusion of an additional phase factor in $C_2(t)$, and the same could be done for phase ϕ if it were nonzero.

The insertion of Equations (33), (34) and (36) into Equation (8) readily gives:

$$\mathbf{v}_t = 2C_1(t)\mathrm{Re}C_2(t)\hat{\mathbf{x}} + 2C_1(t)\mathrm{Im}C_2(t)\hat{\mathbf{y}} + \left[C_1^2(t) - |C_2(t)|^2\right]\hat{\mathbf{z}}. \tag{37}$$

It can be verified that $\mathbf{v}_t$ satisfies the necessary condition $v_t^2 = 1$ at all times.

The fastest dynamic evolution is expected along a great circle arc from the north pole to the south pole of the Bloch sphere [26]. This is compatible with the expression (37) for $\mathbf{v}_t$ if $C_2(t)$ is set as a pure imaginary quantity. Since $U(dt) = 1 - iH(0)dt$ implies that:

$$C_2(dt) = -i\frac{\omega(0)}{2}dt, \tag{38}$$

we write $C_2(t) = -i|C_2(t)|$ and Equation (35) becomes:

$$\mathbf{v}_t = 2C_1(t)|C_2(t)|\hat{\mathbf{y}} + \left[C_1^2(t) - |C_2(t)|^2\right]\hat{\mathbf{z}}. \tag{39}$$

Similarly to the expression (33) for the state of the system, Equation (39) goes beyond the case of CT process. The cross term of the $\mathbf{v}_t$ component along $\hat{\mathbf{y}}$ expresses the interference between the two electronic states necessary to build up the final populations. The $\mathbf{v}_t$ component along $\hat{\mathbf{z}}$ is the population difference between the two diabatic states. By using the normalization condition on the expansion coefficients of the state vector, this component can be rewritten as $2C_1^2(t) - 1$. Considering this expression from the point of view of the occupation of the donor site, rather than from a CT perspective, the $\hat{\mathbf{z}}$ component of $\mathbf{v}_t$ is strictly related to the average charge on the donor molecular site (the same consideration can be applied to the acceptor site), similarly to what is found for charge qubits that correspond to the occupation and de-occupation of a superconducting island by an excess Cooper pair [42].

In CT processes, the cross term describes the transient delocalization of the charge. If the charge finally localizes with unit probability on the acceptor site, this term returns to its initial zero value as the transfer is completed. Accordingly, the population difference passes from 1 (charge initially on the donor site) to −1 (charge finally on the acceptor site).

$\mathbf{v}_t$ describes the aforementioned rotation on the Bloch sphere by setting:

$$\begin{cases} C_1(t) = \cos\kappa(t) = u_0(t) \\ C_2(t) = -i\sin\kappa(t) \end{cases} \tag{40}$$

which implies

$$\mathbf{v}_t = -\sin 2\kappa(t)\hat{\mathbf{y}} + \cos 2\kappa(t)\hat{\mathbf{z}}. \tag{41}$$

Indeed, one can easily see that Equation (40) is the solution of the Schrödinger equation in matrix form for the generic Hamiltonian (25) simplified by choosing $\Omega(t) = 0$ and $\phi = 0$, once $\kappa(t)$ is defined as in Equation (30). The rotation on the Bloch sphere can reach the south pole depending on the form of $\kappa(t)$, that is, on the time dependence of the electronic coupling.

For $t = 0$, Equation (41) gives $\mathbf{v}_0 = \hat{\mathbf{z}}$ (and therefore $v_0^2 = 1$, as expected for a pure state that evolves coherently), which corresponds to the initial CT state, that is, to the north pole on the Bloch sphere. At this point it is readily seen that:

$$\mathbf{v}_t \times \mathbf{v}'_t = 2\kappa'\hat{\mathbf{x}} \tag{42}$$

and

$$\begin{cases} w = 0 \\ \mathbf{v}_0 \cdot \left(\mathbf{v}_t \times \mathbf{v}'_t\right) = 0 \end{cases} \rightarrow \mathbf{B}_\parallel = 0. \tag{43}$$

Finally, using the expression (30) for $\kappa(t)$, one obtains

$$\mathbf{B} = \frac{1}{2}\omega_0(t)\,\hat{\mathbf{x}} \equiv V_{12}(t)\,\hat{\mathbf{x}}. \tag{44}$$

This result can be read in two equivalent ways. In the case considered, the CT corresponds to a rotation on a great circle of the Bloch sphere that is generated by a fictitious magnetic field perpendicular to the orbit whatever the initial point on the sphere, the arc length run, and the travel speed. All these features depend on the specific time-dependent functional form of $\mathbf{B}$. In terms of the Hamiltonian (25), as the states are degenerate and the coupling is real, the state of the system is subject to a rotation which amounts to a changing localization of the charge. In physical–mathematical terms, in cases where the system evolution is unitary and $\mathbf{B}_\parallel = 0$, Equation (18) becomes $\mathbf{B} = \left(\mathbf{v}_t \times \mathbf{v}'_t\right)/2$ and provides the general solution to the dynamical problem, irrespective of the functional form (i.e., whatever the specific initial value and time dependence) of $\mathbf{v}_t$. A specific choice of electronic coupling modulus produces a specific CT dynamics within the general form (40) or (41). For example, $\omega_0(t) = 2\nu\mathrm{sech}(\nu t)$ leads to a smooth increase in the population of the final state, as described by the transition probability $P_{1\rightarrow 2}(t) = |C_2(t)|^2 = \tanh^2(\nu t)$ [8], and the CT rate clearly depends on the coupling.

Now, one can use the formalism in Section 3 to calculate directly the evolution operator. With $u_0(t)$ from Equation (40) and $v_0^2 + \mathbf{v}_0 \cdot \mathbf{v}_t = 1 + \cos 2\kappa(t) = 2\cos^2\kappa(t)$, Equation (23) gives:

$$\mathbf{u}(t) = \frac{\mathbf{v}_0 \times \mathbf{v}_t}{2\cos\kappa(t)} = \frac{\sin 2\kappa(t)}{2\cos\kappa(t)}\hat{\mathbf{x}} = \sin\kappa(t)\hat{\mathbf{x}}, \tag{45}$$

which can be replaced into Equation (3) to obtain:

$$U(t) = \begin{pmatrix} \cos\kappa(t) & -i\sin\kappa(t) \\ -i\sin\kappa(t) & \cos\kappa(t) \end{pmatrix}, \tag{46}$$

consistent with the known result provided by Equation (29) for the present case, as shown in ref. [8]. Note, as a backward comparison, that the system evolution chosen in the inverse problem satisfies Equation (28) with $\Omega(t) = 0$ and $\mu = 0$; ϕ is therefore a constant, which is zero in the above.

6. Concluding Remarks

In this study, we present a general solution to the inverse problem for two-level systems. A sensible point of our theoretical approach is that it starts from expectation values of observables, as is required by experimental measurements.

In the case of a spin-1/2 particle, the magnetic field specifies the Hamiltonian, namely, the system, and therefore its evolution for any given initial conditions. Equation (19) reflects the fact that having two different expectation values of the Pauli operators at times $t \neq 0$ when starting from the same values $\mathbf{v}_0$ at $t = 0$ requires two different driving fields.

The methodology lends itself to applications within more complex systems and generalizations. For example, it was shown that the dynamics of two coupled spin-1/2 systems subject to two different magnetic fields can be reduced to two independent sub-dynamics of the spins [99] that can be exactly solved by appropriate choice of the fields. Therefore, it would be interesting and practically relevant to combine the present approach with that in Ref. [99] to engineer the composite system. This can be particularly useful for applications in quantum computing, where generally a set of qubits is steered from an initial condition to a desired final one. Other generalizations may also be easily enabled, e.g., by the treatment of multilevel spin dynamics as a sequence of two-level transitions at avoided crossings [100]. Finally, the consideration of the spin system in contact with a thermal bath is of clear relevance to the comparison with many experiments and worthy of future analysis. However, it is also worth noting that the charge-transfer model employed describes the single passage through an avoided crossing within the diabatic state framework for a system generally at a nonzero temperature.

We show the meaning and application of the inverse problem solution to CT systems, where the time evolution of the expectation values of the Pauli (or the corresponding spin) operators translates into the interference between the two localized-charge states and their population difference. We emphasize the formal equivalence of the spin in magnetic field and CT systems through the visualization of a CT process on the Bloch sphere. This type of picture has also been used for the coherent excitation of two-level systems, with a different meaning of the physical quantities involved [3,101], and can be useful for quantum information handling purposes, for example to help conceive of information transfer processes that can be accomplished through a given two-state physical system.

We conclude noting that, apart from possible useful extensions, the stand-alone importance of the proposed method is enhanced by the need for coherent control of individual qubits in many quantum technologies [102].

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Appendix A

In this appendix we demonstrate the relationship between $\mathbf{B}$ and $(u_0, \mathbf{u})$ described by Equations (6) and (7). Using the Hamiltonian in Equation (1), the parameterization of the evolution operator in Equation (3), and the definition of the Pauli matrices, we obtain

$$\frac{H}{\hbar}U = \begin{pmatrix} B_3 & B_1 - iB_2 \\ B_1 + iB_2 & -B_3 \end{pmatrix}\begin{pmatrix} u_0 - iu_3 & -u_2 - iu_1 \\ u_2 - iu_1 & u_0 + iu_3 \end{pmatrix} = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix}, \tag{A1}$$

where

$$\begin{aligned} M_{11} &= B_3(u_0 - iu_3) + (B_1 - iB_2)\,(u_2 - iu_1) = B_3u_0 + B_1u_2 - B_2u_1 - i\,\mathbf{B}\cdot\mathbf{u} \\ &= B_3u_0 + (\mathbf{B}\times\mathbf{u})_3 - i\,\mathbf{B}\cdot\mathbf{u}\,, \end{aligned} \tag{A2}$$

$$\begin{aligned} M_{12} &= -B_3(u_2 + iu_1) + (B_1 - iB_2)\,(u_0 + iu_3) \\ &= B_1u_0 + B_2u_3 - B_3u_2 - i(B_2u_0 + B_3u_1 - B_1u_3) \\ &= B_1u_0 + (\mathbf{B}\times\mathbf{u})_1 - i[B_2u_0 + (\mathbf{B}\times\mathbf{u})_2]\,, \end{aligned} \tag{A3}$$

$$\begin{aligned} M_{21} &= (B_1 + B_2)(u_0 - iu_3) - B_3(u_2 - iu_1) \\ &= B_1u_0 + B_2u_3 - B_3u_2 + i(B_2u_0 + B_3u_1 - B_1u_3) \\ &= B_1u_0 + (\mathbf{B}\times\mathbf{u})_1 + i[B_2u_0 + (\mathbf{B}\times\mathbf{u})_2]\,, \end{aligned} \tag{A4}$$

$$\begin{aligned} M_{22} &= -(B_1 + iB_2)(u_2 + iu_1) - B_3(u_0 + iu_3) = -B_3u_0 - (B_1u_2 - B_2u_1) - i\,\mathbf{B}\cdot\mathbf{u} \\ &= -B_3u_0 - (\mathbf{B}\times\mathbf{u})_3 - i\,\mathbf{B}\cdot\mathbf{u}\,. \end{aligned} \tag{A5}$$

Inserting the above quantities and the parameterized expression for $U(t)$ into the Schrödinger equation for the evolution operator, $i\hbar U' = HU$, it is immediately seen that Equation (6) is satisfied. Then, using the vector identity

$$\mathbf{a}\times(\mathbf{b}\times\mathbf{c}) = \mathbf{b}(\mathbf{a}\cdot\mathbf{c}) - \mathbf{c}(\mathbf{a}\cdot\mathbf{b}), \tag{A6}$$

vector multiplication of the second Equation (6) by $\mathbf{u}$ from the left yields

$$\mathbf{u}\times\mathbf{u}' = u_0\mathbf{u}\times\mathbf{B} + \mathbf{u}\times(\mathbf{B}\times\mathbf{u}) = u_0\mathbf{u}\times\mathbf{B} + u^2\mathbf{B} - (\mathbf{u}\cdot\mathbf{B})\mathbf{u}. \tag{A7}$$

Inserting Equation (5) and the second equality (6) into Equation (A7), we obtain

$$\mathbf{u}\times\mathbf{u}' = u_0\left(u_0\mathbf{B} - \mathbf{u}'\right) + u^2\mathbf{B} + u'_0\mathbf{u} = \mathbf{B} + u'_0\mathbf{u} - u_0\mathbf{u}' \;\;\rightarrow\;\; \mathbf{B} = u_0\mathbf{u}' - u'_0\mathbf{u} + \mathbf{u}\times\mathbf{u}', \tag{A8}$$

which is Equation (6).

Appendix B

Inserting Equation (3) for the evolution operator into Equation (8), using the commutators of the Pauli matrices, the relation $\sigma_j\sigma_k = \delta_{jk}1 + i\sum_{l=1}^{3} \varepsilon_{jkl}\sigma_l$ (which is readily obtained by adding the commutator to the anticommutator of the Pauli matrices, and making use of the Kronecker delta and Levi-Civita symbol), the invariance of the trace of a product of operators with respect to cyclic permutations of the operators, and Equation (A6), one obtains Equation (10):

$$\begin{aligned}
\mathbf{v}_t &= \mathrm{Tr}[\boldsymbol{\sigma}(u_0 1 - i\,\mathbf{u}\cdot\boldsymbol{\sigma})\rho_0(u_0 1 + i\,\mathbf{u}\cdot\boldsymbol{\sigma})] \\
&= \mathrm{Tr}[\boldsymbol{\sigma}(u_0\rho_0 u_0 + iu_0\rho_0\mathbf{u}\cdot\boldsymbol{\sigma} - i\,\mathbf{u}\cdot\boldsymbol{\sigma}\rho_0 u_0 + \mathbf{u}\cdot\boldsymbol{\sigma}\rho_0\mathbf{u}\cdot\boldsymbol{\sigma})] \\
&= u_0^2\mathrm{Tr}[\boldsymbol{\sigma}\rho_0] + iu_0\mathrm{Tr}[(\mathbf{u}\cdot\boldsymbol{\sigma})\boldsymbol{\sigma}\rho_0 - \boldsymbol{\sigma}(\mathbf{u}\cdot\boldsymbol{\sigma})\rho_0] + \mathrm{Tr}[(\mathbf{u}\cdot\boldsymbol{\sigma})\boldsymbol{\sigma}(\mathbf{u}\cdot\boldsymbol{\sigma})\rho_0] \\
&= u_0^2\mathbf{v}_0 + iu_0\mathrm{Tr}\left[u_j(\sigma_j\sigma_k - \sigma_k\sigma_j)\hat{\mathbf{c}}_k\rho_0\right] + \mathrm{Tr}\left[u_j\sigma_j\hat{\mathbf{c}}_k\sigma_k u_l\sigma_l\rho_0\right] \\
&= u_0^2\mathbf{v}_0 - 2u_0\mathrm{Tr}\left[\varepsilon_{jkl}\hat{\mathbf{c}}_k\sigma_l u_j\,\rho_0\right] + \hat{\mathbf{c}}_k u_j u_l\mathrm{Tr}\left[\sigma_j\sigma_k\sigma_l\rho_0\right] \\
&= u_0^2\mathbf{v}_0 - 2u_0\varepsilon_{klj}\hat{\mathbf{c}}_k u_j\mathrm{Tr}[\sigma_l\,\rho_0] + \hat{\mathbf{c}}_k u_j u_l\mathrm{Tr}\left[\left(\delta_{jk}1 + i\varepsilon_{jkm}\,\sigma_m\right)\sigma_l\rho_0\right] \\
&= u_0^2\mathbf{v}_0 - 2u_0\varepsilon_{klj}\hat{\mathbf{c}}_k v_{0l}u_j + \hat{\mathbf{c}}_k u_k u_l\mathrm{Tr}[\sigma_l\rho_0] + i\hat{\mathbf{c}}_k u_j u_l\varepsilon_{jkm}\mathrm{Tr}[\sigma_m\sigma_l\rho_0] \\
&= \left(1 - u^2\right)\mathbf{v}_0 + 2u_0\,\mathbf{u}\times\mathbf{v}_0 + \hat{\mathbf{c}}_k u_k u_l v_{0l} + i\hat{\mathbf{c}}_k u_j u_l\varepsilon_{jkl} + \varepsilon_{kjm}\hat{\mathbf{c}}_k u_j\varepsilon_{mln}u_l v_{0n} \\
&= \mathbf{v}_0 + 2u_0\,\mathbf{u}\times\mathbf{v}_0 + \mathbf{u}(\mathbf{u}\cdot\mathbf{v}_0) - \mathbf{v}_0 u^2 + \mathbf{u}\times(\mathbf{u}\times\mathbf{v}_0) \\
&= \mathbf{v}_0 + 2u_0\,\mathbf{u}\times\mathbf{v}_0 + 2\mathbf{u}\times(\mathbf{u}\times\mathbf{v}_0)
\end{aligned} \tag{A9}$$

where Einstein's notation is used for the repeated indices. Using (A6), Equation (A9) is rewritten in the form

$$\mathbf{v}_t - \mathbf{v}_0 = 2u_0\,\mathbf{u}\times\mathbf{v}_0 + 2(\mathbf{u}\cdot\mathbf{v}_0)\mathbf{u} - 2u^2\mathbf{v}_0. \tag{A10}$$

and, using Equation (5),

$$\mathbf{v}_t + \mathbf{v}_0 = 2u_0^2\mathbf{v}_0 + 2u_0\,\mathbf{u}\times\mathbf{v}_0 + 2(\mathbf{u}\cdot\mathbf{v}_0)\mathbf{u}. \tag{A11}$$

By performing the scalar multiplication of (A10) and (A11) side by side, using the properties of the scalar triple product, and denoting θ the angle between $\mathbf{u}$ and $\mathbf{v}_0$, one obtains

$$\begin{aligned}
v_t^2 - v_0^2 &= 4u_0^2u^2v_0^2\sin^2\theta + 4u_0^2u^2v_0^2\cos^2\theta \\
&+ 4u^4v_0^2\cos^2\theta - 4u_0^2u^2v_0^2 - 4u^4v_0^2\cos^2\theta = 0\,,
\end{aligned} \tag{A12}$$

namely, Equation (12). In particular, for a pure state, it is straightforwardly seen using Equation (9) that $v_t^2 = v_0^2 = 1$.

Appendix C

To find the necessary relations between $(u_0, \mathbf{u})$ and $\mathbf{v}_t$, and then the expression for the field, we first use Equations (10), (A6) and (A11) to write the vector product

$$\begin{aligned}
\mathbf{v}_0\times\mathbf{v}_t &= 2u_0\mathbf{v}_0\times(\mathbf{u}\times\mathbf{v}_0) + 2\mathbf{v}_0\times[\mathbf{u}\times(\mathbf{u}\times\mathbf{v}_0)] \\
&= 2\left[u_0v_0^2\mathbf{u} - u_0(\mathbf{u}\cdot\mathbf{v}_0)\mathbf{v}_0 - (\mathbf{u}\cdot\mathbf{v}_0)\,(\mathbf{u}\times\mathbf{v}_0)\right] \\
&= 2u_0v_0^2\mathbf{u} - \tfrac{\mathbf{u}\cdot\mathbf{v}_0}{u_0}(2u_0^2\mathbf{v}_0 + 2u_0\mathbf{u}\times\mathbf{v}_0) \\
&= 2u_0v_0^2\mathbf{u} - \tfrac{\mathbf{u}\cdot\mathbf{v}_0}{u_0}[\mathbf{v}_t + \mathbf{v}_0 - 2(\mathbf{u}\cdot\mathbf{v}_0)\mathbf{u}]
\end{aligned} \tag{A13}$$

whence

$$\mathbf{u} = \frac{(\mathbf{u}\cdot\mathbf{v}_0)(\mathbf{v}_0 + \mathbf{v}_t) + u_0\mathbf{v}_0\times\mathbf{v}_t}{2\left[u_0^2v_0^2 + (\mathbf{u}\cdot\mathbf{v}_0)^2\right]}. \tag{A14}$$

Equation (15) is clearly obtained by dividing both sides of Equation (A14) by u_0, while the scalar multiplication by $\mathbf{v}_0$ produces the following:

$$\begin{aligned}&\mathbf{u}\cdot\mathbf{v}_0=\frac{(\mathbf{u}\cdot\mathbf{v}_0)(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)}{2\left[u_0^2v_0^2+(\mathbf{u}\cdot\mathbf{v}_0)^2\right]}\;\rightarrow\;2\left[u_0^2v_0^2+(\mathbf{u}\cdot\mathbf{v}_0)^2\right]=v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t\\&\rightarrow\;w\equiv\widetilde{\mathbf{u}}\cdot\mathbf{v}_0=\frac{\mathbf{u}\cdot\mathbf{v}_0}{u_0}=\pm\sqrt{\frac{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}{2u_0^2}-v_0^2}\,.\end{aligned}\tag{A15}$$

Equation (16) is thus demonstrated.

Next, we use the vector identity (A6), Equation (12), and the property $\mathbf{v}_t\cdot\mathbf{v}'_t=0$ that results from Equation (13) to write the following expression for $\mathbf{B}$:

$$\begin{aligned}&\frac{\mathbf{B}}{u_0^2}=\hat{\mathbf{u}}'+\hat{\mathbf{u}}\times\hat{\mathbf{u}}'\\&=-\frac{\mathbf{v}_0\cdot\mathbf{v}'_t[w(\mathbf{v}_0+\mathbf{v}_t)+\mathbf{v}_0\times\mathbf{v}_t]}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}+\frac{w'(\mathbf{v}_0+\mathbf{v}_t)+w\mathbf{v}'_t+\mathbf{v}_0\times\mathbf{v}'_t}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}\\&+\frac{w(\mathbf{v}_0+\mathbf{v}_t)+\mathbf{v}_0\times\mathbf{v}_t}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}\times\left\{\frac{w'(\mathbf{v}_0+\mathbf{v}_t)+w\mathbf{v}'_t+\mathbf{v}_0\times\mathbf{v}'_t}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}\right.\\&\left.-\frac{\mathbf{v}_0\cdot\mathbf{v}'_t[w(\mathbf{v}_0+\mathbf{v}_t)+\mathbf{v}_0\times\mathbf{v}_t]}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}\right\}\\&=-\frac{\mathbf{v}_0\cdot\mathbf{v}'_t[w(\mathbf{v}_0+\mathbf{v}_t)+\mathbf{v}_0\times\mathbf{v}_t]}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}+\frac{w'(\mathbf{v}_0+\mathbf{v}_t)+w\mathbf{v}'_t+\mathbf{v}_0\times\mathbf{v}'_t}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}\\&+\frac{1}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}\left[w(\mathbf{v}_0+\mathbf{v}_t)\times(w\mathbf{v}'_t+\mathbf{v}_0\times\mathbf{v}'_t)\right.\\&\left.+(\mathbf{v}_0\times\mathbf{v}_t)\times(w'\mathbf{v}_0+w'\mathbf{v}_t+w\mathbf{v}'_t)+(\mathbf{v}_0\times\mathbf{v}_t)\times(\mathbf{v}_0\times\mathbf{v}'_t)\right]\\&=\frac{1}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}\left\{-(\mathbf{v}_0\cdot\mathbf{v}'_t)\mathbf{v}_0\times\mathbf{v}_t+w^2(\mathbf{v}_0+\mathbf{v}_t)\times\mathbf{v}'_t\right.\\&\left.+(w'\mathbf{v}_t-w\mathbf{v}'_t-w'\mathbf{v}_0)(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)+\left[(\mathbf{v}_t\times\mathbf{v}'_t)\cdot\mathbf{v}_0\right]\mathbf{v}_0\right\}\\&+\frac{w'(\mathbf{v}_0+\mathbf{v}_t)+w\mathbf{v}'_t+\mathbf{v}_0\times\mathbf{v}'_t}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}=\frac{\mathbf{v}_0\cdot(\mathbf{v}_t\times\mathbf{v}'_t)}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}\mathbf{v}_0-\frac{\mathbf{v}_0\cdot\mathbf{v}'_t}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}\mathbf{v}_0\times\mathbf{v}_t\\&+\frac{1}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}\left[2w'\mathbf{v}_t+\left(1+\frac{w^2}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}\right)\mathbf{v}_0\times\mathbf{v}'_t\right]+\frac{w^2\mathbf{v}_t\times\mathbf{v}'_t}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}.\end{aligned}\tag{A16}$$

Singling out the term proportional to $\mathbf{v}_t\times\mathbf{v}'_t$ in Equation (18), we obtain

$$\begin{aligned}&\frac{\mathbf{B}_\parallel}{u_0^2}=\frac{\mathbf{v}_0\cdot(\mathbf{v}_t\times\mathbf{v}'_t)}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}\mathbf{v}_0-\frac{\mathbf{v}_0\cdot\mathbf{v}'_t}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}\mathbf{v}_0\times\mathbf{v}_t\\&+\frac{1}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}\left[2w'\mathbf{v}_t+\left(1+\frac{w^2}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}\right)\mathbf{v}_0\times\mathbf{v}'_t\right]\\&+\left[\frac{w^2}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}-\frac{1}{2u_0^2v_0^2}\right]\mathbf{v}_t\times\mathbf{v}'_t\\&=\frac{2w'\mathbf{v}_t}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}+\frac{\mathbf{v}_0\cdot(\mathbf{v}_t\times\mathbf{v}'_t)\mathbf{v}_0-(\mathbf{v}_0\cdot\mathbf{v}'_t)\mathbf{v}_0\times\mathbf{v}_t+(\mathbf{v}_0\cdot\mathbf{v}_t)\mathbf{v}_0\times\mathbf{v}'_t-v_0^2\mathbf{v}_t\times\mathbf{v}'_t}{(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)^2}\\&+\frac{v_0^2\mathbf{v}_0\times\mathbf{v}'_t-(\mathbf{v}_0\cdot\mathbf{v}_t)\mathbf{v}_t\times\mathbf{v}'_t}{2u_0^2v_0^2(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)}.\end{aligned}\tag{A17}$$

At this point, using the Jacobi identity for the cross product,

$$\begin{aligned}0&=\mathbf{v}_0\times\left[\mathbf{v}_0\times(\mathbf{v}_t\times\mathbf{v}'_t)+\mathbf{v}_t\times(\mathbf{v}'_t\times\mathbf{v}_0)+\mathbf{v}'_t\times(\mathbf{v}_0\times\mathbf{v}_t)\right]\\&=\mathbf{v}_0\cdot(\mathbf{v}_t\times\mathbf{v}'_t)\mathbf{v}_0-v_0^2\mathbf{v}_t\times\mathbf{v}'_t+(\mathbf{v}_0\cdot\mathbf{v}_t)\mathbf{v}_0\times\mathbf{v}'_t-(\mathbf{v}_0\cdot\mathbf{v}'_t)\mathbf{v}_0\times\mathbf{v}_t\,,\end{aligned}\tag{A18}$$

the expression for $\mathbf{B}_\parallel$ reduces to

$$\mathbf{B}_\parallel=\frac{2u_0^2w'\mathbf{v}_t}{v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t}+\frac{v_0^2\mathbf{v}_0\times\mathbf{v}'_t-(\mathbf{v}_0\cdot\mathbf{v}_t)\mathbf{v}_t\times\mathbf{v}'_t}{2v_0^2(v_0^2+\mathbf{v}_0\cdot\mathbf{v}_t)},\tag{A19}$$

which is evidently orthogonal to $\mathbf{v}'_t$, since $\mathbf{v}_t \perp \mathbf{v}'_t$. Furthermore, it is $\mathbf{B}_\parallel \perp \mathbf{v}_t \times \mathbf{v}'_t$. In fact, using the vector identity

$$(\mathbf{a} \times \mathbf{b}) \cdot (\mathbf{c} \times \mathbf{d}) = (\mathbf{a} \cdot \mathbf{c})(\mathbf{b} \cdot \mathbf{d}) - (\mathbf{a} \cdot \mathbf{d})(\mathbf{b} \cdot \mathbf{c}) \tag{A20}$$

and Equation (12), it is obtained:

$$\left[v_0^2(\mathbf{v}_0 \times \mathbf{v}'_t) - (\mathbf{v}_0 \cdot \mathbf{v}_t)(\mathbf{v}_t \times \mathbf{v}'_t)\right] \cdot (\mathbf{v} \times \mathbf{v}'_t) = v_0^2(\mathbf{v}_0 \cdot \mathbf{v}_t)v_t'^2 - (\mathbf{v}_0 \cdot \mathbf{v}_t)v_t^2 v_t'^2 = 0. \tag{A21}$$

Since $\mathbf{B}_\parallel \perp \{\mathbf{v}'_t, \mathbf{v}_t \times \mathbf{v}'_t\}$, it is very easy to see geometrically that $\mathbf{B}_\parallel$ is parallel to $\mathbf{v}_t$. To show this analytically, we make use of the expansion of $\mathbf{v}_0$ on the orthogonal vector basis $\{\mathbf{v}_t, \mathbf{v}'_t, \mathbf{v}_t \times \mathbf{v}'_t\}$, which, using Equation (12) and the fact that $\mathbf{v}_t \perp \mathbf{v}'_t$, is written

$$\mathbf{v}_0 = \frac{\mathbf{v}_0 \cdot \mathbf{v}_t}{v_0^2}\mathbf{v}_t + \frac{\mathbf{v}_0 \cdot \mathbf{v}'_t}{v_t'^2}\mathbf{v}'_t + \frac{\mathbf{v}_0 \cdot (\mathbf{v}_t \times \mathbf{v}'_t)}{v_0^2 v_t'^2}\mathbf{v}_t \times \mathbf{v}'_t. \tag{A22}$$

Inserting this expression into the product $\mathbf{v}_0 \times \mathbf{v}'_t$ of Equation (A19) gives

$$\begin{aligned} \mathbf{B}_\parallel &= \tfrac{2u_0^2 w' \mathbf{v}_t}{v_0^2 + \mathbf{v}_0 \cdot \mathbf{v}_t} - \tfrac{\mathbf{v}_0 \cdot (\mathbf{v}_t \times \mathbf{v}'_t)}{2v_0^2 v_t'^2 (v_0^2 + \mathbf{v}_0 \cdot \mathbf{v}_t)}\mathbf{v}'_t \times (\mathbf{v}_t \times \mathbf{v}'_t) \\ &= \left[\tfrac{2u_0^2 w'}{v_0^2 + \mathbf{v}_0 \cdot \mathbf{v}_t} - \tfrac{\mathbf{v}_0 \cdot (\mathbf{v}_t \times \mathbf{v}'_t)}{2v_0^2 (v_0^2 + \mathbf{v}_0 \cdot \mathbf{v}_t)}\right]\mathbf{v}_t, \end{aligned} \tag{A23}$$

namely, Equation (19).

Appendix D

In this Appendix, we derive the expression (20) for the time autocorrelation of the Pauli vector and the related Equation (23). Using the identity $\varepsilon_{ljk}\,\varepsilon_{mjk} = 2\delta_{lm}$, the properties of the scalar triple product, and Equation (5), one obtains

$$\begin{aligned} &\mathcal{C}(t) = \mathrm{Tr}[(u_0 1 + i\,\mathbf{u} \cdot \boldsymbol{\sigma})\boldsymbol{\sigma}(u_0 1 - i\,\mathbf{u} \cdot \boldsymbol{\sigma}) \cdot \boldsymbol{\sigma}\rho_0] \\ &= u_0^2 \mathrm{Tr}[\boldsymbol{\sigma} \cdot \boldsymbol{\sigma}\rho_0] + iu_0 \mathrm{Tr}\{[(\mathbf{u} \cdot \boldsymbol{\sigma})\boldsymbol{\sigma} - \boldsymbol{\sigma}(\mathbf{u} \cdot \boldsymbol{\sigma})] \cdot \boldsymbol{\sigma}\rho_0\} + \mathrm{Tr}\{[(\mathbf{u} \cdot \boldsymbol{\sigma})\boldsymbol{\sigma}(\mathbf{u} \cdot \boldsymbol{\sigma})] \cdot \boldsymbol{\sigma}\rho_0\} \\ &= 3u_0^2 \mathrm{Tr}[\rho_0] + iu_0 \mathrm{Tr}\left[u_j(\sigma_j\sigma_k - \sigma_k\sigma_j)\sigma_l\delta_{kl}\rho_0\right] + \mathrm{Tr}\left[u_j\sigma_j\sigma_k u_l\sigma_l\sigma_m\delta_{km}\rho_0\right] \\ &= 3u_0^2 - 2u_0 \mathrm{Tr}\left[u_j\varepsilon_{jkm}\,\sigma_m\sigma_k\rho_0\right] + u_j u_l \mathrm{Tr}\left[\sigma_j\sigma_k(\sigma_k\sigma_l + 2i\varepsilon_{lkm}\,\sigma_m)\rho_0\right] \\ &= 3u_0^2 - 2u_0 \mathrm{Tr}\left[u_j\varepsilon_{jkm}\,(\delta_{mk}1 + i\varepsilon_{mkn}\,\sigma_n)\rho_0\right] + 3u_j u_l \mathrm{Tr}\left[\sigma_j\sigma_l\rho_0\right] \\ &+2iu_j\varepsilon_{lkm}\,u_l \mathrm{Tr}\left[\sigma_j\sigma_k\sigma_m\rho_0\right] = 3u_0^2 + 2iu_0 \mathrm{Tr}\left[u_j\varepsilon_{jkm}\,\varepsilon_{nkm}\,\sigma_n\rho_0\right] \\ &+3u_j u_k \mathrm{Tr}\left[\left(\delta_{jk}1 + i\varepsilon_{jkl}\,\sigma_l\right)\rho_0\right] + 2iu_j\varepsilon_{lkm}\,u_l \mathrm{Tr}\left[\sigma_j(\delta_{km}1 + i\varepsilon_{kmn}\,\sigma_n)\rho_0\right] \\ &= 3u_0^2 + 4iu_0 \mathrm{Tr}\left[u_j\delta_{jn}\,\sigma_n\rho_0\right] + 3u^2 + 3iu_j\varepsilon_{jkl}\,u_k v_{0l} \\ &-2u_j u_l\varepsilon_{\,lkm}\,\varepsilon_{nkm}\,\mathrm{Tr}\left[\left(\delta_{jn}1 + i\varepsilon_{jnp}\,\sigma_p\right)\rho_0\right] \\ &= 3u_0^2 + 4iu_0 u_j \mathrm{Tr}\left[\sigma_j\rho_0\right] + 3u^2 - 4u_j u_l\delta_{ln}\delta_{jn} - 4iu_j u_l\varepsilon_{jnp}\,\delta_{ln} \mathrm{Tr}\left[\sigma_p\rho_0\right] \\ &= 3u_0^2 + 4iu_0 u_j v_{0j} + 3u^2 - 4u^2 - 4iu_j\varepsilon_{jkl}\,u_k\,v_{0l} = 4u_0^2 - 1 + 4iu_0(\mathbf{u} \cdot \mathbf{v}_0) \end{aligned} \tag{A24}$$

where Einstein's notation is used again for the summation over the repeated indices. Equation (A24) gives Equation (20), whence (21) and (22).

To derive Equation (23), we first note that the insertion of Equation (22) into the third equality (A15) gives

$$\mathbf{u} \cdot \mathbf{v}_0 = \pm\sqrt{\frac{v_0^2 + \mathbf{v}_0 \cdot \mathbf{v}_t}{2} - \frac{v_0^2}{4}[1 + \mathrm{Re}\mathcal{C}(t)]}. \tag{A25}$$

Equation (23) readily results from using Equations (22) and (A25) in the numerator of the expression (A14) for **u** and the second equality (A15) in its denominator.

Appendix E

Here, we derive the general expression (24) for the fundamental two-observable time correlations of Pauli operators. Using Equation (5), Einstein's notation for the repeated indices, and the identity $\varepsilon_{jkl}\varepsilon_{jmn} = \delta_{km}\delta_{ln} - \delta_{kn}\delta_{lm}$, one finds

$$\begin{aligned}
&\mathcal{C}_{np}(t) = \mathrm{Tr}\left[(u_0 1 + i\,\mathbf{u}\cdot\boldsymbol{\sigma})\sigma_n(u_0 1 - i\,\mathbf{u}\cdot\boldsymbol{\sigma})\sigma_p\rho_0\right] \\
&= u_0^2\mathrm{Tr}\left[\sigma_n\sigma_p\rho_0\right] + iu_0\mathrm{Tr}\{[(\mathbf{u}\cdot\boldsymbol{\sigma})\sigma_n - \sigma_n(\mathbf{u}\cdot\boldsymbol{\sigma})]\sigma_p\rho_0\} \\
&+\mathrm{Tr}\{[(\mathbf{u}\cdot\boldsymbol{\sigma})\sigma_n(\mathbf{u}\cdot\boldsymbol{\sigma})]\sigma_p\rho_0\} \\
&= u_0^2\mathrm{Tr}\left[(\delta_{np}1 + i\varepsilon_{npq}\,\sigma_q)\rho_0\right] + iu_0\mathrm{Tr}\left[u_j(\sigma_j\sigma_n - \sigma_n\sigma_j)\sigma_p\rho_0\right] \\
&+\mathrm{Tr}\left[u_j\sigma_j\sigma_n u_l\sigma_l\sigma_p\rho_0\right] = u_0^2\delta_{np} + iu_0^2\varepsilon_{npq}v_{0q} - 2u_0\mathrm{Tr}\left[u_j\varepsilon_{jnk}\,\sigma_k\sigma_p\rho_0\right] \\
&+u_ju_l\mathrm{Tr}\left[\left(\delta_{jn}1 + i\varepsilon_{jnk}\,\sigma_k\right)\left(\delta_{lp}1 + i\varepsilon_{lpm}\,\sigma_m\right)\rho_0\right] = u_0^2(\delta_{np} + i\varepsilon_{npq}v_{0q}) \\
&-2u_0\mathrm{Tr}\left[u_j\varepsilon_{jnk}\left(\delta_{kp}1 + i\varepsilon_{kpq}\,\sigma_q\right)\rho_0\right] + u_nu_p + iu_ju_l\varepsilon_{lpm}\,\delta_{jn}\mathrm{Tr}[\sigma_m\rho_0] \\
&+iu_ju_l\varepsilon_{jnk}\,\delta_{lp}\mathrm{Tr}[\sigma_k\rho_0] - u_ju_l\varepsilon_{jnk}\varepsilon_{lpm}\mathrm{Tr}\left[\left(\delta_{km}1 + i\varepsilon_{kmq}\,\sigma_q\right)\rho_0\right] \\
&= u_0^2(\delta_{np} + i\varepsilon_{npq}v_{0q}) - 2u_0u_j\left[\varepsilon_{jnk}\,\delta_{kp} + i(\delta_{jp}\delta_{nq} - \delta_{jq}\delta_{np})v_{0q}\right] + u_nu_p \\
&-iu_n\varepsilon_{plm}\,u_lv_{0m} - iu_p\varepsilon_{njk}\,u_jv_{0k} - u_ju_l\varepsilon_{jnk}\varepsilon_{lpk} - iu_ju_l\varepsilon_{jnk}\left(\delta_{lq}\delta_{kp} - \delta_{lp}\delta_{kq}\right)v_{0q} \\
&= u_0^2(\delta_{np} + i\varepsilon_{npq}v_{0q}) + 2u_0(\mathbf{u}\times\hat{\mathbf{c}}_p)_n - 2iu_0v_{0n}u_p + 2iu_0u_qv_{0q}\delta_{np} + u_nu_p \\
&-iu_n(\mathbf{u}\times\mathbf{v}_0)_p - iu_p(\mathbf{u}\times\mathbf{v}_0)_n - u_ju_l\left(\delta_{jl}\delta_{np} - \delta_{jp}\delta_{ln}\right) - i\varepsilon_{jnp}u_ju_qv_{0q} - i\varepsilon_{njq}u_jv_{0q}u_p \\
&= (u_0^2 + 2iu_0\mathbf{u}\cdot\mathbf{v}_0 - u^2)\delta_{np} + [2u_n - 2iu_0v_{0n} - i(\mathbf{u}\times\mathbf{v}_0)_n]u_p \\
&+\{[(2u_0 + i\mathbf{u}\cdot\mathbf{v}_0)\mathbf{u} - iu_0^2\mathbf{v}_0]\times\hat{\mathbf{c}}_p\}_n - iu_n(\mathbf{u}\times\mathbf{v}_0)_p - iu_p(\mathbf{u}\times\mathbf{v}_0)_n \\
&= 2\left(u_0^2 + iu_0\mathbf{u}\cdot\mathbf{v}_0 - \tfrac{1}{2}\right)\delta_{np} + 2[u_n - iu_0v_{0n} - i(\mathbf{u}\times\mathbf{v}_0)_n]u_p \\
&+\{[(2u_0 + i\mathbf{u}\cdot\mathbf{v}_0)\mathbf{u} - iu_0^2\mathbf{v}_0]\times\hat{\mathbf{c}}_p\}_n - iu_n(\mathbf{u}\times\mathbf{v}_0)_p
\end{aligned} \tag{A26}$$

from which Equation (24) results.

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