Quantum Theory of Radical-Ion-Pair Recombination: A New Physical Paradigm for Low-Magnetic-Field Effects

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A new paradigm emerging in the description of magnetic-sensitive radical-ion-pair recombination reactions is presented. This paradigm is founded on the realization that the recombination process of radical-ion pairs is a continuous quantum measurement. In the regime of low magnetic fields we describe the appearance of purely quantum phenomena, that fade away as the magnetic field increases. We will analyze the magnetic sensitivity of these reactions under this perspective and bridge the new full quantum theory with the existing classical reaction theory applicable at high magnetic fields. Based on the quantum theory of recombination we will then explain experimental observations incompatible with classical reaction theory, in particular the effect of deuteration on the magnetic sensitivity of radical-ion pair recombination yields.

I. INTRODUCTION

Radical-ion pairs have been at the focus of chemical, biological and physical research for the last few decades, mostly because of their relevance to photosynthesis [1]. It is a chain of electron-transfer reactions [2] that converts the photon energy absorbed by pigment molecules in the photosynthetic reaction center to energy capable of driving further chemical reactions of biological importance. The radical-ion pairs formed in this intricate photochemical machinery further complicate the picture, since the spin interactions of their unpaired electrons make the whole process magnetic-sensitive. The importance of the magnetic sensitivity of photosynthetic reactions [3, 4, 5] was realized early on and intensely studied theoretically. The radical-ion pairs formed in this intricate photochemical reaction center to energy capable of driving further chemical reactions of biological importance.

Parallel to these developments, it was early on realized [6, 7] that the magnetic sensitivity of radical-ion pair (RIP) recombination reactions might be at the basis of the biochemical magnetic compass used by several species for navigation in earth’s magnetic field [8]. After the development of extensive theoretical models [9, 10, 11, 12] and several supporting experiments [13, 14, 15], the so-called RIP magnetoreception mechanism is now established as forming the basis of the biochemical compass, at least of migratory avian species.

The magnetic sensitivity of RIP reactions [16] can be easily understood: the RIP is formed from a photoexcited donor-acceptor dyad molecule (DA) in the singlet spin state. Magnetic interactions with the external magnetic field and hyperfine interactions with the molecule’s nuclear spins induce the so-called singlet-triplet (S-T) mixing. Since the recombination of the RIP is spin-selective, i.e. the singlet S(D+ A−) RIP can recombine to the neutral DA molecule, whereas the triplet T(D+ A−) combines, if at all, to a triplet metastable excited state TDA, it is seen that the magnetic field can directly influence the end-result of the photoexcitation-recombination cycle.

What has gone unnoticed so far, however, is the fact that the spin-selective charge recombination process is fundamentally a quantum measurement [17]. The tunneling of the RIP into a vibrational excited state of the DA molecule is a scattering process the outcome of which depends on the RIP’s spin state. In other words, the intermolecular recombination dynamics continuously interrogate the radical-ion pair’s spin state. This interrogation can be readily described by quantum measurement theory [18] and leads to purely quantum phenomena such as the quantum Zeno effect [19]. The theoretical description of magnetic-sensitive RIP recombination processes was so far based on phenomenological classical reaction theory [20] which masked the quantum effects that are dominant in the regime of low-magnetic fields. This is the reason that classical theories were rather successful, i.e. most measurements were performed at high magnetic fields, where classical reaction theories are valid. Several puzzling experimental observations at low fields were however left unexplained. As will be shown in the following, the quantum Zeno effect crucially affects the relevant time constants of the RIP recombination process, and naturally leads to an understanding of several experimental findings that could not be reconciled with classical RIP reaction theories. In this article we will present the detailed quantum theory of the RIP recombination process and elucidate the fundamentally different interpretation of the low-magnetic-field effects that emerges. We will outline the connection of the full quantum theory to be presented with the classical reaction theories. We will also provide an explanation of experimental findings that have challenged the classical theory, in particular measurements regarding the effect of deutera-
II. THE QUANTUM DESCRIPTION OF RIP RECOMBINATION

In Figure 1 we diagrammatically describe the evolution of the RIP recombination. The RIP is at time \( t = 0 \) created in the singlet state \( S(D^+A^-) \), which coherently mixes with the triplet state \( T(D^+A^-) \) under the unitary action of the magnetic Hamiltonian \( \mathcal{H} \). At the same time, the charge recombination is performing a quantum measurement of the singlet- and triplet-state projection operators \( Q_S \) and \( Q_T \), with the measurement rate being \( k_S \) and \( k_T \), respectively. At some point the measurement is over. If the measurement result is \( q_S = 1 \) (obviously meaning that \( q_T = 0 \)), the RIP can tunnel to an excited vibrational state of the neutral DA molecule, \( S(DA^+)^* \), and from there decay to the DA ground state at a rate \( \gamma_S \). In the event that the measurement produces the result \( q_S = 0 \) (obviously meaning that \( q_T = 1 \)), the RIP similarly ends up at a metastable triplet state of the DA molecule, \( T(DA^+)^* \), and this happens at a rate \( \gamma_T \). In this work we will not deal with the decay rates \( \gamma_S \) and \( \gamma_T \), since they not are relevant for the magnetic dynamics.

Quantum measurement theory [18] describes the effect of a continuous measurement of a system observable \( q \) performed at a rate \( k \) on the system’s time evolution by the Liouville equation:

\[
\frac{d\rho}{dt} = -i[\mathcal{H}, \rho] - k[q, [q, \rho]]
\]  

(1)

where \( \rho \) is the quantum system’s density matrix and the first term describes the unitary evolution due to a Hamiltonian interaction \( \mathcal{H} \). In the case of the RIP recombination process, the measured system observables are \( Q_S \) and \( Q_T \). Therefore, the RIP’s density matrix evolution equation will be

\[
\frac{d\rho}{dt} = -i[\mathcal{H}, \rho] - k_S[Q_S, [Q_S, \rho]] - k_T[Q_T, [Q_T, \rho]]
\]  

(2)

Measurements performed on a coherently evolving quantum system always cause decoherence. That is, (2) describes the evolution of an initially pure state of the RIP to a mixed state, during which the coherence induced by the Hamiltonian \( \mathcal{H} \) is also dissipated. This is due to the measurement-induced back-action. The RIP magnetic Hamiltonian to be considered is comprised of the Zeeman interaction of the two unpaired electrons with the external magnetic field and by the hyperfine interactions with the molecule’s nuclear magnetic moments.

We will consider for simplicity only two nuclei, with nuclear spins \( I_1 \) and \( I_2 \), and different hyperfine couplings (in frequency units) \( A_1 \) and \( A_2 \) to each of the electrons. We will consider isotropic hyperfine interactions, as we are not interested in describing directional effects [21] in this work. The magnetic Hamiltonian thus reads

\[
\mathcal{H} = h_1 + h_2
\]  

(3)

where

\[
h_{ij} = \omega s_{jz} + a_j I_j \cdot s_j
\]  

(4)

is the interaction Hamiltonian for each electron and \( j = 1, 2 \). The Larmor frequency is

\[
\omega = g_s \mu_B B = 2.8 \frac{\text{MHz}}{G} B[G]
\]  

(5)

and throughout we are using units such that \( \hbar = 1 \). For earth’s field of \( \sim 0.5 \text{ G} \), \( \omega = 0.7 \mu \text{s}^{-1} \). In the following, \( \omega \) will be the unit of frequency, i.e. we will normalize all rates by \( \omega \). The normalization is performed by \( \omega \) and not by the hyperfine frequency scale \( a_1 \) (or \( a_2 \)) for the following reason. If the mechanism is to serve for the measurement of the magnetic field \( \omega \), with precision \( \delta \omega \), then from Heisenberg’s time-energy uncertainty relation follows that in a measurement time \( \tau \), the achievable precision \( \delta \omega \) is such that \( \tau \delta \omega \sim 1 \). The precision \( \delta \omega \) is understood to be the width of the measured distribution of frequencies \( \omega \). Since the measurement time \( \tau \) is determined by the decay rate \( \lambda \) of the RIP S-T mixing, \( \tau \sim 1/\lambda \), the relative width of the measured distribution is \( \delta \omega/\omega \sim \lambda/\omega \). Therefore Heisenberg’s uncertainty poses the requirement that \( \lambda/\omega < 1 \), if the mechanism is
to be sensitive to the magnetic field itself. If however, the signal-to-noise of the actual detection process is \( s \), than the measurement sensitivity is enhanced to \( \lambda/\omega \). For example, if the magnetic-field change is detected by the change in the recombined molecules yield and the latter can be measured at the level of \( 1\% \), then \( \delta\omega/\omega \sim 0.01 \) if \( \lambda/\omega \sim 1 \). It is obvious that the condition \( \lambda/\omega < 1 \) can be more easily satisfied at high fields, since the classical recombination rates \( k \) (and hence the decay rates \( \lambda \)) are usually much larger than \( 1 \mu s^{-1} \). In the following, we are going to show how the quantum Zeno effect allows the existence of long S-T lifetimes, so that the condition \( \lambda/\omega \sim 1 \) is satisfied even if \( k \gg 1 \mu s^{-1} \).

We will now derive some basic properties of (2). We denote by \( S = \text{Tr}\{\rho Q_S\} \) and \( T = \text{Tr}\{\rho Q_T\} \) the probability to find the RIP in the singlet and triplet spin state, respectively. Due to the fact that \( Q_S \) and \( Q_T \) are orthogonal projection operators, we have \( Q_S^2 = Q_S \), \( Q_T^2 = Q_T \), \( Q_S + Q_T = 1 \). Therefore, using the identity \( \text{Tr}\{[A,B]\} = 0 \), it follows from (2) that

\[
\frac{dS}{dt} = -i\text{Tr}\{[Q_S, \mathcal{H}]\rho\} \tag{6}
\]

\[
\frac{dT}{dt} = -i\text{Tr}\{[Q_T, \mathcal{H}]\rho\} \tag{7}
\]

and

\[
\text{Tr}\{\rho\} = S + T = 1 \tag{8}
\]

Does the fact that \( \text{Tr}\{\rho\} = 1 \) mean that the RIP never recombines? Not at all. Every time the RIP’s spin state approaches the point \( S = 1 \) (or \( T = 1 \)), there is a non-zero probability of singlet- or triplet channel charge recombination. If the RIP recombines, it ceases to exist in the Hilbert space defined by the eigenstates of \( \mathcal{H} \), and together with is ceases the meaning of the density matrix \( \rho \) and its normalization. This is formally described by a quantum stochastic process \( 22, 23 \) taking place in an enlarged Hilbert space. A brief analysis has appeared in \( 17 \) and a more detailed presentation of the RIP’s spin state evolution will be presented in a forthcoming manuscript. Such a description requires the complete solution of the density matrix equation (2). In the following we will present some general arguments that do not necessitate such a solution.

III. EIGENVALUE SPECTRUM OF THE LIOUVILLE EQUATION

Since the nuclear spin multiplicity of the problem we are considering is \( (2I_1 + 1)(2I_2 + 1) \), and the electron spin multiplicity is 4, the density matrix is a square \( N \)-dimensional matrix, where \( N = 4(2I_1+1)(2I_2+1) \). Equation (2) thus represents a set of \( N^2 \) coupled first-order differential equations. We can rewrite them in the form \( dR/dt = AR \), where \( R = (\rho_{11}, \rho_{12} ... \rho_{NN})^T \) is a column vector of dimension \( N^2 \) containing all density matrix elements and \( A \) is a \( N^2 \)-dimensional square matrix. The eigenvalues of \( A \) are playing a central role in the considerations of this work, since they are governing the time evolution of the RIP’s spin state. These eigenvalues are found by numerically diagonalizing the matrix \( A \), and in general are complex numbers of the form \( -\lambda + i\Omega \), where \( \lambda \geq 0 \) is termed the decay rate and \( \Omega \) the mixing frequency of this particular eigen-mode. There are \( n = N^2 \) such eigenmodes. For example, if \( I_1 = 1/2 \) and \( I_2 = 1 \), then \( n = 576 \). It is easily seen that \( n \) grows exponentially with the number of participating spins, and the numerical diagonalization quickly becomes a formidable problem. This is the reason we limit the calculations to the case of just two nuclear spins.

A result of such a diagonalization is shown in Figure 2 for typical values of the problem’s parameters. We assume that \( k_T < k_S \), so that the recombination rate scale is determined by \( k_S \). We will distinguish two physically distinct regimes: (i) \( k_S/\omega \ll 1 \), and (ii) \( k_S/\omega \gg 1 \). We term the first regime classical, since it is in this case that classical reaction theories are valid, whereas the second is termed quantum, because it is in this parameter regime that the quantum Zeno effect is at play. Before examining the magnetic sensitivity in these two regimes, we note the following. Since the time evolution of the density matrix is described by \( n \) eigenmodes, every density matrix element can be written as

\[
\rho_{ij}(t) = \sum_{l=1}^{n} c^{(l)}_{ij} e^{(-\lambda_l t + i\Omega_l t)}, \tag{9}
\]

where the coefficients \( c^{(l)}_{ij} \) are determined from the initial...
conditions. Therefore

$$
\langle Q_S(t) \rangle = \sum_{ij} \rho_{ij}(t) q_{ji} = \sum_{l=1}^{n} e^{(-\lambda_l + iQ_l)t} A_l
$$

(10)

where $\rho_{ij} = \langle i|\rho|j \rangle$ and $|i\rangle$ with $i = 1, ..., n$ are the basis states, and

$$
A_l = \sum_{ij} e^{iQ_l} q_{ji}
$$

(11)

with $q_{ji} = \langle j|Q_S|i \rangle$. The reality of $\langle Q_S(t) \rangle$ is based on the appearance of positive and negative mixing frequencies, as shown in Figure 2B.

A. Classical Regime

We here study the magnetic sensitivity of RIP reactions when $k_S/\omega < 1$, i.e. in the regime of small recombination rates and/or large magnetic fields. This is termed the classical regime. We first note that in this regime  the decay rates $\lambda$ scale proportionally to $k_S$, $\lambda \sim k_S$. Secondly, the mixing frequencies are constant, i.e. independent of the recombination rate $k_S$. Hence all $n$ terms contribute to $\langle Q_S(t) \rangle$, i.e. $n_{slow} = n$ in (12). In this case, the magnetic sensitivity of $\langle Q_S(t) \rangle$ stems from the dependence of $q_{ji}$ on $\omega$, i.e. a change in $\omega$ will result in a change of $A_l$. As can be seen from Figure 3B, the matrix elements $q_{ij}$ and hence $A_l$ are varying strongly for fields $\omega/a > 1$ until the very high field regime $\omega/a \gg 1$, where the magnetic sensitivity drops to zero. In this high-field regime, the $T_\pm$ branches of the energy eigenvalues have separated away and there is small mixing only between $T_0$ and $S$, as can be seen in Figure 3A. The above considerations form the classical interpretation of magnetic field effects on RIP recombination reactions, that have already been analyzed in detail [8, 10] (and references therein).

B. Quantum Regime

The situation changes dramatically as the ratio $k_S/\omega$ increases. Indeed, it is seen from Figure 3C that for low magnetic fields, $\omega/a \ll 1$, a 10% change in $\omega$ produces a mere 2% change in $q_{ij}$. According to classical theory, this is further suppressed by the ratio $\omega/k_S$, resulting in a negligible change of $\langle Q_S(t) \rangle$. However, significant recombination yield changes are indeed observed experimentally. The reason is that in this regime quantum effects become dominant, fundamentally altering the nature of the magnetic sensitivity of the recombination process. As seen in Figure 2A, it is in this regime that the decay rates split into two branches, the fast (upper branch) and slow (lower branch) decay rates. The latter are a manifestation of the quantum Zeno effect [19], the physical significance of has been already described elsewhere [20]. We will here elaborate on the new interpretation of magnetic sensitivity of RIP reactions in this quantum regime. Let us suppose that some of the decay rates $\lambda_l$ are slow, i.e. $\lambda_l \ll \Omega_l$, and some other are fast, i.e. $\lambda_l \gg \Omega_l$. Let us further suppose that there are $n_{slow}$ of the former and hence $n_{fast} = n - n_{slow}$ of the latter. Thus $n_{fast}$ terms in (10) will quickly decay away, leaving the $n_{slow}$ terms to determine the long-time behavior of $\langle Q_S(t) \rangle$:

$$
\langle Q_S(t) \rangle_{t \gg 1/\lambda_{fast}} = \sum_{l=1}^{n_{slow}} e^{(-\lambda_l + i\Omega_l)t} A_l
$$

(12)

It thus follows that in this regime it is mainly the change of the number $n_{slow}$ with the magnetic field $\omega$ that is
FIG. 4: (A) Decay rates only the lower branch is shown) as a function of $\omega$ for $k_S = 15$, $k_T = 0.2k_S$, nuclear spins $I_1 = I_2 = 1/2$ and hyperfine couplings $a_1 = 1.5$ and $a_2 = 3$. The dashed red line is the $\lambda = \omega$ line. The magnetic sensitivity derives from the changing number of decay rates crossing this line.

responsible for the magnetic sensitivity of $\langle Q_S(t) \rangle$. This number is a strongly changing function of $\omega$ as shown in Figure 4.

IV. PHENOMENOLOGICAL LIOUVILLE EQUATIONS

The phenomenological density matrix evolution equations used so far are

$$\frac{d\rho}{dt} = -i[H, \rho] - k_S(Q_S\rho + \rho Q_S) - k_T(Q_T\rho + \rho Q_T)$$

(13)

The decay rates of the eigenvalue spectrum of (13) are shown in Figure 5. It is seen that all the decay rates scale in proportion to $k_S$, i.e. the quantum Zeno effect has disappeared. This is expected, because equation (13) by design forces the normalization of the density matrix, $\text{Tr}\{\rho\}$, to an exponential decay:

$$\frac{d\text{Tr}\{\rho\}}{dt} = -k_S\langle Q_S \rangle - k_T\langle Q_T \rangle$$

(14)

There is an analogous situation in atomic physics experiments: transit time broadening [24]. When an atomic coherence (for example a ground state spin coherence) of an atomic vapor is probed by a laser, the atoms stay in the interaction volume (defined by the laser beam) for a limited time $\tau_{tr}$, determined by the atomic velocity distribution. The measured spin coherence lifetime will thus be limited by $\tau_{tr}$, which will add to the spin-resonance width by $1/\tau_{tr}$. This is the transit-time broadening, which would be described by an equation similar to (14):  

$$\frac{d\text{Tr}\{\rho\}}{dt} = -\frac{\text{Tr}\{\rho\}}{\tau_{tr}}$$

(15)

This equation phenomenologically describes the disappearance of atoms with a time constant $\tau_{tr}$. The situation with radical-ion pairs, however, is fundamentally different, since in this case there is a selective quantum measurement continuously going on. An attempt to artificially introduce the crucial term $Q_S\rho Q_S$ existing in [13] and missing from (13) was made in [25] based on phenomenological considerations, without any further consequences on the theoretical description of RIP recombination dynamics. All theoretical treatments, summarized in [20], have thereafter used the phenomenological density matrix equation (13).

V. EXPLANATION OF EXPERIMENTAL OBSERVATIONS INCONSISTENT WITH CLASSICAL REACTION THEORY

We will now apply the interpretation of the low-field magnetic sensitivity of RIP recombination reaction previously put forward to address experimental observations that seem paradoxical within the framework of classical reaction theory. Unusually long time constants that have been observed in RIP reactions several times have been explained elsewhere [17]. Recently observed [15] magnetic sensitivity of RIP reaction rates which is in stark contrast with classical reaction theory has been shown [27] to be fully consistent with the quantum theory. Several experimental observations relevant to avian magnetoreception based on magnetic-sensitive RIP reactions have also been accounted for [26]. Here we focus on some other puzzling observations, namely the effect of the RIP’s deuteration on the magnetic sensitivity of the RIP recombination.
A. Magnetic Sensitivity and Deuteration

The effects of deuteration on the magnetic sensitivity of RIP recombination reactions have early on been noted as signaling the inadequacies of classical reaction theories, as has been noted in [28]. As stated in [28], "The identical triplet quantum yields in H and D samples does not support the generally accepted idea that hyperfine interactions are responsible for spin rephasing in the P870+I- radical pair". Indeed, it is rather straightforward to derive, based on the classical spin-state mixing description, the result of a change of hyperfine couplings. The triplet recombination yield is crudely expected [29] to scale as $a/k_S$, where $a$ is the dominant hyperfine coupling and $k_S$ the singlet recombination rate. Any change in $a$ should thus lead to a proportional change in the yield, contrary to the 1979 observations.

It is rather straightforward to explain this apparent contradiction by use of the eigenvalue spectrum of the quantum evolution equation [2]. Indeed, if we calculate the eigenvalue spectrum for a molecule with $I_1 = I_2 = 1/2$ and hyperfine couplings $a_1 = 1.5\omega$ and $a_2 = 2.8a_1$, we find that in the quantum regime ($k_S/\omega \gg 1$) the number of decay rates $\lambda$ such that $\lambda/\omega < 1$ are 58.2% of the total (256). If we repeat the calculation for $I_1 = 1/2$, $I_2 = 1$, $a_1 = 1.5\omega$ and $a_2 = 0.86a_1$, i.e. we replaced the second nucleus, which before was assumed to be a hydrogen, with a deuterium, we find that the previous percentage changes to 62.5% (the total number of eigenvalues is now 576) i.e. the relative yield change is only 7%. However, the change in the hyperfine coupling is about 300%, clearly exemplifying the disparity of classical expectations with observed data. We will now focus on more recent data regarding the effect deuteration has on the magnetic sensitivity [30, 31] of the the pyrene (Py)-dimethylaniline (DMA) RIP recombination dynamics. What has been observed is that the low-field-effect is most pronounced when only one of the radicals exhibits strong hyperfine couplings, with the second radical being deuterated and hence having substantially reduced hyperfine couplings (the magnetic moment of deuterium is about one third of the proton’s). In particular, in the case of protonated pyrene RIPs Py-h$_{10}$/DMA-h$_{11}^+$ and Py-h$_{10}$/DMA-d$_{11}^+$, there was almost no magnetic-sensitivity of the exciplex fluorescence. On the contrary, the recombination dynamics of deuterated pyrene RIPs Py-d$_{10}$/DMA-h$_{11}^+$ and Py-d$_{10}$/DMA-d$_{11}^+$ showed a clear low-field sensitivity. In Figure 6 we show the calculated behavior of the magnetic sensitivity. For each of the four RIPs we calculate the eigenvalue spectrum and count the decay rates $\lambda$ that are smaller than $\omega$. This spectrum is shown for one of the RIPs in Figure 6A. Half the percentage of those decay rates is a good estimate [26] for the decrease in the singlet recombination yield. It is seen that the relative yield change with magnetic field displayed in Figure 6B is in perfect agreement with observations. The absolute values of the yield drop are not accurately predicted due to the simplicity of the two-spin model used (they also depend on the particular value of $k_S$).

In summary, we have analyzed the physical interpretation of low magnetic field effects that is bestowed upon the radical-ion pair recombination reactions by their full quantum-mechanical description. The explanatory power of this theory has been applied in understanding experimental observations on the magnetic sensitivity of RIP reactions that were conflicting classical reaction theories. It has been shown that the eigenvalue spectrum of the density matrix equation describing the time evolution of the RIP’s spin state accounts for the observations regarding RIP deuteration. In particular, the slow decay rates imposed by the quantum Zeno effect on the eigenvalue spectrum, and their dependence on the problem’s
physical parameters, are shown to contain rich dynamical information. Finally, it is rather crucial to address the effect of low magnetic fields on photosynthetic reactions under this new perspective.

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