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Rotating wave approximation: systematic expansion and application to coupled spin pairs

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Abstract. We propose a new treatment of the dynamics of a periodically time-dependent Liouvillian by mapping it onto a time-independent problem and give a systematic expansion for its effective Liouvillian. In the case of a two-level system, the lowest order contribution is equivalent to the well-known rotating wave approximation. We extend the formalism to a pair of coupled two-level systems. For this pair, we find two Rabi frequencies and we can give parameter regimes where the leading order of the expansion is suppressed and higher orders become important. These results might help to investigate the interaction of tunneling systems in mixed crystals by providing a tool for the analysis of echo experiments.

PACS. 76.60.Lz Spin echoes – 66.35.+a Quantum tunneling of defects

1 Introduction

Quantum tunneling of substitutional defect ions in alkali halide crystals leads to particular low temperature properties [1,2]. Due to their misfit in size or shape such defect ions are confined to a potential energy landscape with a few degenerate potential wells. At low temperatures thermally activated hopping is inhibited and the defect ion passes through the barrier by quantum tunneling. These tunneling defects are usually modeled by two-level-systems (TLSs), which dynamically are identical to spin degrees of freedom.

Depending on the detailed nature of the TLS, both electromagnetic and acoustic fields might couple to the system. One of the interesting features of such systems is their response to an oscillating weak external field which is in resonance with the TLS. The key point in this context is the emergence of the Rabi oscillation [3], which is related to the amplitude of the external field and which allows a systematic experimental investigation of the response of a TLS.

The easiest way to derive the Rabi oscillation is to do the so called *rotating wave approximation* (RWA) as originally introduced by Rabi [4]. Since then, the problem has been tackled by numerous methods starting with the work of Bloch and Siegert [5], who could first give a correction to the RWA. Further approaches have been a solution in terms of continued fractions presented by Autler and Townes [6] and especially the Floquet formalism developed by Shirley [7].

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In this paper, we present yet another expansion scheme for solving the problem. It is somewhat related to the Floquet formalism, but we try to find an effective static Liouvillian or Hamiltonian rather than a time-evolution operator, which makes our calculations easier. Like the Floquet formalism, our approach can be easily generalised to systems involving several energy levels, where the simple geometrical interpretation of the RWA fails. Thus, we can define the RWA in these situations as the lowest order contribution of our expansion scheme.

Our paper is organised as follows. In the following section, we address the time-dependent problem of a single spin. We first rederive the RWA, then in Section 2.2, we present the formalism for our expansion. In the third section then, we use our new method for treating the problem of two coupled spins. The main results of this section are the emergence of two Rabi frequencies, whose relative magnitudes depend on the spin-spin coupling, and the identification of parameter regimes where higher order corrections to the RWA become important.

2 The treatment of a single spin

This section deals with the problem of one TLS that is coupled to an external force. The TLS is modeled by the Hamiltonian [1,2]

$$H_{\rm sys} = -k\frac{\sigma_x}{2} - \Delta\frac{\sigma_z}{2}, \qquad (1)$$

with tunneling element k, asymmetry Δ, and energy $\epsilon = \sqrt{k^2+\Delta^2}$. The external force is given by

$$H_{\rm ext} = -2\eta\,\sin(\omega t)\frac{\sigma_z}{2}\,\cdot \qquad (2)$$

This choice of $H_{\rm ext}$ implies that the external field couples solely to the spatial coordinate σ_z of the TLS, as realized in the case of an electric field coupling to the dipole moment of the TLS.

The time evolution of the statistical operator is governed by the Bloch equation [3,8]

$$\frac{{\rm d}\rho}{{\rm d}t} = {\rm i}[\rho, H_{\rm sys}+H_{\rm ext}] - \gamma(\rho-\rho^{\rm eq}) =: \mathcal{L}\rho, \qquad (3)$$

where the right hand side of the equation defines the Liouvillian $\mathcal{L}$. The second term in the Bloch equation describes a relaxation of the statistical operator towards its equilibrium given by $\rho^{\rm eq} = {\rm e}^{-\beta H_{\rm sys}}/\left({\rm tr}\ {\rm e}^{-\beta H_{\rm sys}}\right)$. This choice of $\rho^{\rm eq}$ is justified as long as the relaxation time is much longer than the period of the driving field, $\gamma \ll \omega$. If we expand the density matrix in terms of the Pauli matrices and the $\mathbf{1}$-matrix,

$$\rho = \frac{1}{2}(\mathbf{1} + r_x\sigma_x + r_y\sigma_y + r_z\sigma_z) =: \begin{pmatrix}1\\ r_x\\ r_y\\ r_z\end{pmatrix}, \qquad r_x^2+r_y^2+r_z^2 \le 1, \qquad (4)$$

the Liouvillian $\mathcal{L}$ can be represented by a 4×4 matrix. Equation (3) then yields a differential equation for the vector $(1, r_x, r_y, r_z)$.

The Hamiltonian $H_{\rm sys}+H_{\rm ext}$ describes not only a tunneling system coupled to a harmonic oscillator, but equally well a spin$-\frac{1}{2}$ particle subjected to a static magnetic field $\mathbf{B} = (k/(g\mu_{\rm B}), 0, \Delta/(g\mu_{\rm B}))$, and an oscillating field in z-direction, $B_z = 2\eta/(g\mu_{\rm B})\sin(\omega t)$. In the latter interpretation, the components of the density matrix (r_x, r_y, r_z) are proportional to the magnetic moment of the spin. Thus, under rotation they transform like a pseudo-vector. In the following we will use the terminologies of both interpretations.

As a preliminary, we consider the case of a static magnetic field. From the classical analogue one can infer that the magnetic moment will precess round the direction of the magnetic field with a frequency proportional to the field strength. Thus, the static Hamiltonian

$$H = \epsilon\left(e_x\frac{\sigma_x}{2} + e_y\frac{\sigma_y}{2} + e_z\frac{\sigma_z}{2}\right), \quad e_x^2+e_y^2+e_z^2 = 1 \qquad (5)$$

describes a rotation of the vector (r_x, r_y, r_z) about the axis (e_x, e_y, e_z) with an angular frequency ϵ. The explicit formula for the statistical operator as a function of time is useful in the following, we give it for $\gamma = 0$, *i.e.* for the case without dissipation: $\rho(t) = \mathcal{U}_0(t)\rho(0) = {\rm e}^{-{\rm i}Ht}\rho(0){\rm e}^{{\rm i}Ht}$, where the time evolution operator $\mathcal{U}_0(t)$ can be represented by the matrix

$$\mathcal{U}_0(t) = \begin{pmatrix}1 & 0 & 0 & 0\\ 0 & c+e_x^2(1-c) & e_xe_y(1-c)-e_zs & e_xe_z(1-c)+e_ys\\ 0 & e_xe_y(1-c)+e_zs & c+e_y^2(1-c) & e_ye_z(1-c)-e_xs\\ 0 & e_xe_z(1-c)-e_ys & e_ye_z(1-c)+e_xs & c+e_z^2(1-c)\end{pmatrix}, \qquad (6)$$

using the abbreviations $c := \cos(\epsilon\, t),\ s := \sin(\epsilon\, t)$.

In the presence of dissipation, the corresponding solution reads

$$\rho(t) = \mathcal{U}(t)\rho(0) = \rho^{\rm st} + {\rm e}^{-\gamma t}\,\mathcal{U}_0(t)\big(\rho(0)-\rho^{\rm st}\big)\,, \qquad (7)$$

where the stationary solution $\rho^{\rm st}$ is given by $\mathcal{L}\,\rho^{\rm st} = 0$.

2.1 RWA

Let us first rederive the RWA. To simplify the problem, we neglect the asymmetry of $H_{\rm sys}$. Furthermore, we assume throughout this section that both the coupling to the external field and the damping are small and that we are close to resonance, *i.e.*

$$\eta, \gamma, |\epsilon-\omega| \ll \omega. \qquad (8)$$

First, we split the Hamiltonian into two parts

$$H = \underbrace{-\omega\,\frac{\sigma_x}{2}}_{H_0}\underbrace{-\delta\,\frac{\sigma_x}{2} - 2\eta\,\sin(\omega t)\,\frac{\sigma_z}{2}}_{H_1} \qquad \text{with} \qquad \delta := \epsilon-\omega, \qquad (9)$$

where assumption (8) ensures that H_1 is a weak perturbation to H_0.

The dominant part H_0 of the Hamiltonian leads to a rotation of the magnetic moment about the x-axis. We take this into account by changing to a rotating frame of reference. In this rotating reference frame, we denote the statistical operator by $\bar\rho$. It is related to ρ through

$$\rho(t) = {\rm e}^{-{\rm i}H_0t}\bar\rho(t){\rm e}^{{\rm i}H_0t} = {\rm e}^{{\rm i}\omega t(\sigma_x/2)}\bar\rho(t){\rm e}^{-{\rm i}\omega t(\sigma_x/2)} \qquad (10)$$

and its equation of motion reads $\frac{{\rm d}\bar\rho}{{\rm d}t} = {\rm i}[\bar\rho, \bar H] - \gamma(\bar\rho-\rho^{\rm eq})$ with the transformed Hamiltonian

$$\bar H = -\delta\,\frac{\sigma_x}{2} + \eta\,\frac{\sigma_y}{2} - \eta\,{\rm e}^{-{\rm i}2\omega t(\sigma_x/2)}\,\frac{\sigma_y}{2}\,{\rm e}^{{\rm i}2\omega t(\sigma_x/2)}. \qquad (11)$$

The form of the effective Hamiltonian is easily understood by observing that the linearly polarised field $H_{\rm ext}$ can be decomposed into two counterrotating circularly polarised fields of strength $(B_z/2)$ and angular frequency ω each. In the rotating frame of reference, one of them becomes a static field whereas the frequency of the other one is doubled.

In the following we want to neglect the oscillating field. This is justified by the following consideration. During one period of the oscillating field, $\bar\rho$ changes only little, as its typical time constant is $1/(\sqrt{\delta^2+\eta^2}) \gg (1/2\omega)$. Therefore, it is plausible to assume that the contributions of the time dependent field cancel over a period. This approximation is called *rotating wave approximation* because the original sinusoidal magnetic field is replaced by a rotating field of half the amplitude and the same frequency. The remaining part of the Hamiltonian is static,

$$\bar{H}_{\text{eff}} = -\delta\,\frac{\sigma_x}{2} + \eta\,\frac{\sigma_y}{2}, \tag{12}$$

describing a rotation with the *Rabi frequency* $\varOmega := \sqrt{\delta^2+\eta^2}$. Thus, we can solve the equations of motion using (7) while the stationary solution reads

$$\begin{aligned}\rho^{\text{st}} &= \frac{1}{2}\Big[\mathbf{1} + \frac{n^0}{\delta^2+\eta^2+\gamma^2}\Big\{(\delta^2+\gamma^2)\,\sigma_x \\ &\quad - \eta\delta\,\sigma_y - \eta\gamma\,\sigma_z\Big\}\Big], \\ n^0 &= \tanh(\beta\epsilon/2).\end{aligned} \tag{13}$$

If at $t=0$, the system is in equilibrium, we find for the dipole moment of the TLS in the laboratory frame

$$\begin{aligned}\langle\hat{p}\rangle(t) &= \text{tr}(\hat{p}\rho(t)) \\ &= -n^0\frac{\eta}{\tilde{\varOmega}}\Big[\cos(\omega t)\Big(\frac{\gamma}{\tilde{\varOmega}} + \mathrm{e}^{-\gamma t}\Big\{\frac{\varOmega}{\tilde{\varOmega}}\sin(\varOmega t) \\ &\quad - \frac{\gamma}{\tilde{\varOmega}}\cos(\varOmega t)\Big\}\Big) + \sin(\omega t) \\ &\quad \times\left(-\frac{\delta}{\tilde{\varOmega}} + \mathrm{e}^{-\gamma t}\left\{\frac{\delta}{\tilde{\varOmega}}\cos(\varOmega t) + \frac{\delta\gamma}{\varOmega\tilde{\varOmega}}\sin(\varOmega t)\right\}\right)\Big]\end{aligned} \tag{14}$$

using the definition $\tilde{\varOmega}^2 := \varOmega^2+\gamma^2$. Fourier transformation yields peaks at ω and $\omega\pm\varOmega$ whose heights have an overall factor of $(\eta/\tilde{\varOmega})$, ensuring that we only find a signal close to resonance.

Although the method of the rotating wave approximation is intuitively very appealing, it lacks a rigorous foundation. In particular, there are no means to estimate the quality of the approximation and its limits of validity.

2.2 An expansion scheme for the RWA

In this section, we present a systematic expansion, which yields the rotating wave approximation as the lowest order contribution. We consider the Hamiltonian

$$H = -k\frac{\sigma_x}{2} - \varDelta\frac{\sigma_z}{2} - 2\eta\sin(\omega t)\frac{\sigma_z}{2}, \tag{15}$$

where only the strength of the driving field η and the damping constant of the Bloch equation γ are small compared to ω:

$$\eta, \gamma \ll \omega. \tag{16}$$

The time evolution is dominated by the static part of the Hamiltonian. Therefore we start by diagonalizing this part by means of a rotation about the y-axis,

$$\begin{aligned}\hat{H} &= \mathrm{e}^{-\mathrm{i}\phi(\sigma_y/2)}\,H\,\mathrm{e}^{\mathrm{i}\phi(\sigma_y/2)} \\ &= -\epsilon\frac{\sigma_x}{2} - 2u\eta\,\sin(\omega t)\frac{\sigma_z}{2} - 2v\eta\,\sin(\omega t)\frac{\sigma_x}{2},\end{aligned} \tag{17}$$

where

$$\epsilon = \sqrt{k^2+\varDelta^2}, \qquad \phi = \arctan(k/\varDelta),$$
$$u = \cos(\phi), \qquad v = \sin(\phi).$$

After this unitary transformation, the equilibrium value of the statistical operator reads

$$\rho^{\text{eq}} = \frac{1}{2}\left[\mathbf{1} + n^0\sigma_x\right], \quad n^0 = \tanh(\beta\epsilon/2). \tag{18}$$

Now, we proceed in a similar way as in the last section. The Hamiltonian is divided into two parts

$$\begin{aligned}\hat{H} &= \underbrace{-n\omega\,\frac{\sigma_x}{2}}_{H_0}\underbrace{-\delta\,\frac{\sigma_x}{2} - 2u\eta\,\sin(\omega t)\frac{\sigma_z}{2} - 2v\eta\,\sin(\omega t)\frac{\sigma_x}{2}}_{H_1}, \\ \delta &:= \epsilon - n\omega,\end{aligned} \tag{19}$$

where the integer n has to be chosen such that $|\delta| < \omega/2$ thereby ensuring that H_1 is small compared to ω. The effect of H_0 can be taken into account by changing to a frame of reference spinning around the x-axis with the frequency $|n\omega|$

$$\begin{aligned}\rho(t) &= \mathrm{e}^{-\mathrm{i}H_0t}\bar{\rho}(t)\mathrm{e}^{\mathrm{i}H_0t} = \mathrm{e}^{\mathrm{i}n\omega t(\sigma_x/2)}\bar{\rho}(t)\mathrm{e}^{-\mathrm{i}n\omega t(\sigma_x/2)} \\ \frac{\mathrm{d}\bar{\rho}}{\mathrm{d}t} &= \mathrm{i}[\bar{\rho},\bar{H}] - \gamma(\bar{\rho}-\rho^{\text{eq}}) =: \bar{\mathcal{L}}\bar{\rho},\end{aligned} \tag{20}$$

where $\bar{\mathcal{L}}$ reads in the basis $(1, \sigma_x, \sigma_y, \sigma_z)$

$$\bar{\mathcal{L}}(t) = \begin{pmatrix} 0 & 0 & 0 & 0 \\ \gamma n^0 & -\gamma & -u\eta\,g(t) & u\eta\,f(t) \\ 0 & u\eta\,g(t) & -\gamma & \delta + 2v\eta\,\sin(\omega t) \\ 0 & -u\eta\,f(t) & -(\delta+2v\eta\,\sin(\omega t)) & -\gamma \end{pmatrix}, \tag{21}$$

using the definitions

$$\begin{aligned}f(t) &= \cos((n-1)\,\omega t) - \cos((n+1)\,\omega t) \\ g(t) &= \sin((n-1)\,\omega t) - \sin((n+1)\,\omega t).\end{aligned}$$

In the following, we first show that there is an effective, static Liouville operator yielding a time evolution that matches the correct time evolution at integer multiples of the period $\tau := 2\pi/\omega$. Then, we give an expansion of the effective Liouville operator in powers of $(1/\omega)$.

If we use the time ordering operator $\mathcal{T}$, the formal solution for the evolution of $\bar{\rho}$ reads

$$\bar{\rho}(t) = \mathcal{T}\exp\left[\int_0^t \bar{\mathcal{L}}(t')\,\mathrm{d}t'\right]\bar{\rho}(0). \tag{22}$$

Taking advantage of the periodicity of $\bar{\mathcal{L}}(t)$, we rewrite the last equation as

$$\bar{\rho}(t) = \left\{\mathcal{T}\exp\left[\int_{N\tau}^t \bar{\mathcal{L}}(t')\mathrm{d}t'\right]\right\}\left\{\mathcal{T}\exp\left[\int_0^\tau \bar{\mathcal{L}}(t')\mathrm{d}t'\right]\right\}^N \bar{\rho}(0),$$
$$N\tau < t < (N+1)\tau.$$

Now, we define the effective Liouvillian $\mathcal{L}_{\rm eff}$ by

$$\exp\left[\mathcal{L}_{\rm eff}\tau\right] = \mathcal{T}\exp\left[\int_0^\tau \bar{\mathcal{L}}(t')\,{\rm d}t'\right] \tag{23}$$

and end up with

$$\bar{\rho}(t) = \left\{\mathcal{T}\exp\left[\int_{N\tau}^t \bar{\mathcal{L}}(t')\,{\rm d}t'\right]\right\}\exp\left[\mathcal{L}_{\rm eff}N\tau\right]\bar{\rho}(0). \tag{24}$$

Thus, we separated the problem into two parts: first, one has to calculate the evolution between $N\tau$ and t accessible by simple perturbation theory. Second, one has to determine $\mathcal{L}_{\rm eff}$. We discuss the latter first.

We use the identity

$$\exp[\mathcal{L}_{\rm eff}N\tau] = \mathcal{T}\exp\left[\int_0^{N\tau}\bar{\mathcal{L}}(t'){\rm d}t'\right]$$

$$\Longleftrightarrow\quad \mathcal{L}_{\rm eff}N\tau + \frac{1}{2}\mathcal{L}_{\rm eff}^2(N\tau)^2 + \ldots = \int_0^{N\tau}{\rm d}t_1\,\bar{\mathcal{L}}(t_1) + \int_0^{N\tau}{\rm d}t_1\int_0^{t_1}{\rm d}t_2\,\bar{\mathcal{L}}(t_1)\bar{\mathcal{L}}(t_2) + \ldots, \tag{25}$$

which implies that $\mathcal{L}_{\rm eff}$ is the sum of all terms on the right hand side proportional to $N\tau$. Luckily, these terms form a series in $(1/\omega)$ that can be calculated explicitly order by order. By substituting $z_i := \omega t_i$, the mth integral yields

$$\frac{1}{\omega^m}\int_0^{2\pi N}{\rm d}z_1\int_0^{z_1}{\rm d}z_2\ldots\int_0^{z_{m-1}}{\rm d}z_m\bar{\mathcal{L}}(z_1/\omega)\bar{\mathcal{L}}(z_2/\omega)\ldots\bar{\mathcal{L}}(z_m/\omega) = \frac{1}{\omega^m}\sum_{i=1}^m C_i(2\pi N)^i, \tag{26}$$

where the coefficients C_i do not depend on ω. Only the terms proportional to N contribute to $\mathcal{L}_{\rm eff}$ and we obtain

$$\mathcal{L}_{\rm eff}^{(m-1)} = \frac{1}{\omega^{m-1}}\,C_1 \tag{27}$$

at order $m-1$. Our choice of δ and the condition (16) assure that we can truncate the series after a few terms.

To lowest order, we have

$$\mathcal{L}_{\rm eff}^{(0)} = \frac{1}{N\tau}\int_0^{N\tau}{\rm d}t_1\,\bar{\mathcal{L}}(t_1).$$

In this integral, all oscillating terms vanish. So, to lowest order the driving field contributes only at resonance ($n = 1$), where we obtain the results of the rotating wave approximation (see Sect. 2.1). A calculation of the first order corrections in the case of resonance ($n = 1$) yields

$$\mathcal{L}_{\rm eff} = \mathcal{L}_{\rm eff}^{(0)} + \mathcal{L}_{\rm eff}^{(1)}$$
$$\mathcal{L}_{\rm eff}^{(1)} = \frac{\eta}{\omega}\begin{pmatrix} 0 & 0 & 0 & 0\\ 0 & 0 & -2uv\eta & -\frac{1}{2}u\delta\\ \frac{1}{2}\gamma u n^0 & 2uv\eta & 0 & \frac{3}{4}u^2\eta\\ 0 & \frac{1}{2}u\delta & -\frac{3}{4}u^2\eta & 0\end{pmatrix}. \tag{28}$$

This can be expressed in terms of an effective Hamiltonian and an effective equilibrium state defined by the relation

$$\mathcal{L}_{\rm eff}\bar{\rho} =: {\rm i}[\bar{\rho}, H_{\rm eff}] - \gamma(\bar{\rho} - \rho_{\rm eff}^{\rm eq}). \tag{29}$$

We obtain up to first order

$$H_{\rm eff} = \left(-\delta - \frac{\eta}{\omega}\frac{3u^2\eta}{4}\right)\frac{\sigma_x}{2} + \left(u\eta - \frac{\eta}{\omega}\frac{u\delta}{2}\right)\frac{\sigma_y}{2} + \frac{\eta}{\omega}2uv\eta\,\frac{\sigma_z}{2} \tag{30}$$

and

$$\rho_{\rm eff}^{\rm eq} = \rho^{\rm eq} + \frac{\eta}{\omega}\frac{1}{2}un^0\frac{\sigma_y}{2}\,. \tag{31}$$

Thus we see that the resonance is shifted to $\epsilon = \omega - \frac{\eta}{\omega}\frac{3u^2\eta}{4}$.

If $n \neq 1$, the effective Hamiltonian reads

$$H_{\rm eff} = \left(-\delta - \frac{\eta}{\omega}\frac{n\eta u^2}{n^2-1}\right)\frac{\sigma_x}{2} + \frac{\eta}{\omega}\frac{2u\delta}{n^2-1}\,\frac{\sigma_y}{2} - \delta_{n,2}\,\frac{\eta}{\omega}\eta uv\,\frac{\sigma_z}{2} + \mathcal{O}(\omega^{-2}), \tag{32}$$

while the σ_z-term appears only for $n = 2$. This term yields a resonance with the Rabi frequency $\frac{\eta}{\omega}\eta uv$ when the first term vanishes, *i.e.* at $\epsilon = 2\omega - \frac{\eta}{\omega}\frac{2\eta u^2}{3}$. Similarly, for every n there is a term of order $(\eta/\omega)^{n-1}$ resulting in a resonance at $\epsilon \approx n\omega$, which can be interpreted as the absorption of n field quanta [7,9]. In the symmetric case ($v = 0$), all resonances for even n vanish as can be seen explicitly for $n = 2$ above.

We still have to calculate the evolution between $N\tau$ and t. This can be done using perturbation theory. With $\mathcal{L}_1(t) := \bar{\mathcal{L}}(t) - \mathcal{L}_{\rm eff}$, we write the time evolution operator as the series

$$\begin{aligned}\mathcal{U}(t) &= \mathcal{U}(t-N\tau)\,\mathcal{U}(N\tau)\\ &= {\rm e}^{\mathcal{L}_{\rm eff}t} + \int_{N\tau}^t{\rm d}t_1\,{\rm e}^{\mathcal{L}_{\rm eff}(t-t_1)}\mathcal{L}_1(t_1){\rm e}^{\mathcal{L}_{\rm eff}t_1}\\ &\quad + \int_{N\tau}^t{\rm d}t_1\int_{N\tau}^{t_1}{\rm d}t_2\,{\rm e}^{\mathcal{L}_{\rm eff}(t-t_1)}\mathcal{L}_1(t_1){\rm e}^{\mathcal{L}_{\rm eff}(t_1-t_2)}\\ &\quad \times\mathcal{L}_1(t_2){\rm e}^{\mathcal{L}_{\rm eff}t_2} + \ldots\end{aligned} \tag{33}$$

Thus, we see that to lowest order the time evolution is completely described by the effective Liouvillian, while the corrections are small due to our choice of δ and the condition (16).

In the first part of this paper, we presented a formalism in which we treated a harmonically driven TLS. We could map the time dependent problem onto a static one and give, order by order, an effective Liouvillian describing it. The lowest order result could be identified with the RWA.

This formalism is not restricted to a simple TLS, but can be easily generalised to any system driven by a periodic field. Thus we can generalise the RWA to systems that have not a simple geometrical interpretation like the TLS by identifying it with the lowest order effective Liouvillian. In the second part, we demonstrate this by investigating a coupled pair of TLSs in a harmonic field.

3 A pair of coupled tunneling systems

Depending on the detailed nature of the TLSs in mixed crystals, an electric and an elastic moment is connected to the defect which can thus interact with lattice vibrations, external fields or neighbouring defects. Consequently only at low defect concentrations one can describe the situation by isolated TLSs. With rising concentration pairs, triples etc. of defects are involved until finally one faces a complicated many-body system [2,10].

Echo experiments [3] are an experimental tool to investigate such systems [11]. In the following we investigate the influence of the interaction between the tunneling defects in such experiments. Since we treat pairs of TLSs our results are confined to the limited range where the concentration is sufficiently high to give a considerable amount of pairs but still below the value where many-body effects set in.

We model a pair of coupled tunneling systems (TLSs) by the Hamiltonian

$$H_{\mathrm{pair}} = -\frac{k+\delta k}{2}\sigma_x \otimes 1 - \frac{k-\delta k}{2} 1 \otimes \sigma_x - J\sigma_z \otimes \sigma_z. \tag{34}$$

The first and second term describe symmetric TLSs with $k > \delta k > 0$, while the third term models the dipole-dipole-interaction of the TLSs with the coupling constant J. The external harmonic force, coupling to dipole moment p of the pair, is represented by

$$H_{\mathrm{ext}} = -\eta \sin(\omega t)(\sigma_z \otimes 1 + 1 \otimes \sigma_z) \tag{35}$$

with the external frequency ω and the coupling constant η. This ansatz assumes that the two TLSs are parallel to each other and to the harmonic force. The full Hamiltonian of the problem reads $H = H_{\mathrm{pair}} + H_{\mathrm{ext}}$ and we are interested in the time-dependent dipole moment of the pair assuming that we start from thermal equilibrium.

In the following, we outline the calculations, while details can be found in Appendix A.4. First, we diagonalize H_{pair} to find two *uncoupled effective* TLSs:

$$\hat{H}_{\mathrm{pair}} = -\frac{\epsilon_1}{2}\sigma_x \otimes 1 - \frac{\epsilon_2}{2} 1 \otimes \sigma_x, \tag{36}$$

with the energies $\epsilon_{1/2} = \sqrt{J^2+k^2} \pm \sqrt{J^2+\delta k^2}$. These effective TLSs have a more complicated coupling to the external field than the original ones, reading

$$\hat{H}_{\mathrm{ext}} = -\eta \sin(\omega t)\left\{(u_1 + u_2\sigma_x) \otimes \sigma_z + \sigma_z \otimes (u_3 + u_4\sigma_x)\right\} \tag{37}$$

where the u's dependend on k, δk and J with values in the interval $[-1, 1]$ (for further information see Appendix A.1).

Now, we take the dominant effect of $\hat{H}_{\mathrm{pair}}$ into account by changing to a rotating frame of reference

$$\hat{\rho}(t) = \exp(\mathrm{i}\omega t(n\sigma_x \otimes 1 + m 1 \otimes \sigma_x)/2)\tilde{\rho}(t) \times \exp(-\mathrm{i}\omega t(n\sigma_x \otimes 1 + m 1 \otimes \sigma_x)/2) \tag{38}$$

where the integers n and m have to be chosen such that

$$|\epsilon_1 - n\omega| < \frac{1}{2}\omega \quad \text{and} \quad |\epsilon_2 - m\omega| < \frac{1}{2}\omega.$$

Regarding only resonance phenomena we have to distinguish three cases. The first two cases describe a situation where the internal frequencies ϵ_1 and ϵ_2 are clearly separated. In the first case, the external frequency ω is in resonance with the smaller internal frequency ϵ_2 ($m = 1$ and $n > 1$), while in the second case it is in resonance with ϵ_1 ($m = 0$ and $n = 1$). In the third case, ω is close to both internal frequencies ($m = n = 1$). This implies that both the energy separation δk and the coupling J of the underlying TLSs are small compared to their energy k, *i.e.* J, $\delta k \ll k$.

We will discuss mainly the first case with $m = 1$ and $n > 1$. The results for the second case are essentially the same and can be found in Appendix A.3. The treatment of the third case has also been relegated to the appendix; in spite of more complicated calculations it gives no principally new insight into the problem.

Neglecting dissipation ($\gamma = 0$), focusing on the situation of exact resonance ($\epsilon_2 = \omega$) and following Section 2.2, one obtains for the dipole moment to lowest order:

$$p^{(0)}(t) = \frac{1}{4}\sum_{i=1,2} \frac{\Omega_i}{\eta} A_i \left\{\sin((\omega - \Omega_i)t) - \sin((\omega + \Omega_i)t)\right\} \tag{39}$$

with

$$\begin{aligned} \Omega_{1/2} &= \eta(u_1 \pm u_2) \\ A_{1/2} &= r_2(1 \pm r_1) \\ r_{1/2} &= \tanh(\beta\epsilon_{1/2}/2). \end{aligned} \tag{40}$$

Thus we find *two* Rabi frequencies $\Omega_{1/2}$ yielding four frequencies for the dipole moment: $\omega_p = \epsilon_2 \pm \Omega_1$ and $\omega_p = \epsilon_2 \pm \Omega_2$. This is one of the main results of our paper. Although the two TLSs can be transformed into effective *uncoupled* TLSs they do not give the same results as truly uncoupled TLSs, which have only *one* Rabi frequency. In Figure 1 we have plotted the two Rabi frequencies (Eq. (40)) *versus* the coupling constant J. At $J = 0$, *i.e.* if the two spins are not coupled, the two Rabi frequencies coincide, thus reproducing the result of a single spin. For increasing J, Ω_1 grows, tending quickly to 2η, whereas Ω_2 goes down to zero. In the case of negative J, Ω_1 and Ω_2 interchange their roles.

The amplitudes of the peaks (Eq. (40)) are proportional to the frequencies themselves, thereby enhancing the signal of the bigger Rabi frequency. Additionally the amplitudes also contain a temperature dependent factor $A_{1/2}$ shown in Figure 2. Over the whole temperature range, A_1 is larger than A_2. This is most pronounced for very low temperatures where A_1 tends to a value of 2 whereas A_2 vanishes. For higher temperatures starting at about $T = \epsilon_1$ they are of the same order.

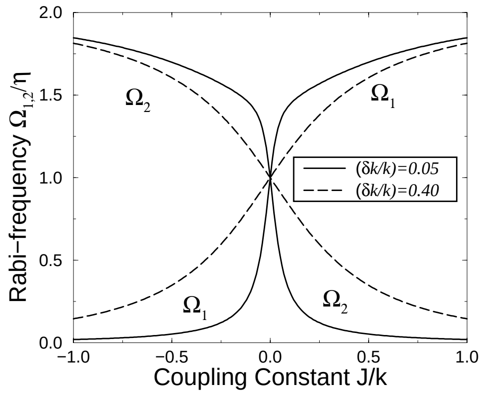

Fig. 1. The two Rabi frequencies $\Omega_{1,2}$ for two different values of $(\delta k/k)$ against (J/k).

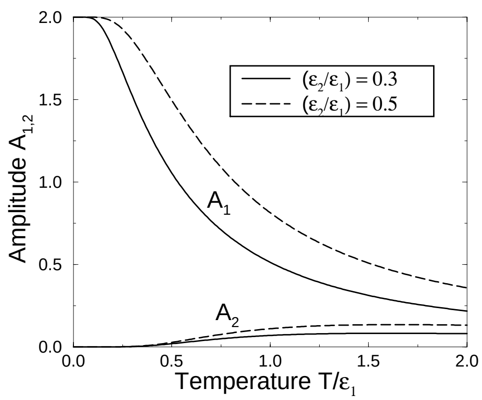

Fig. 2. $A_{1/2} = r_2(1 \pm r_1)$ for two different values of (ϵ_2/ϵ_1) against (T/ϵ_1).

For $J > 0$, A_1 corresponds to the higher Rabi frequency. In this case the higher Rabi frequency peak dominates the spectrum for all temperatures, reaching its maximum for small temperature, while the contribution of the lower Rabi frequency can be seen only at medium temperatures. In the strong coupling limit the lower Rabi frequency and thus also its amplitude vanish, leaving us with only one Rabi frequency of 2η. This is in accordance with the picture that two strongly coupled TLSs form one small *effective* TLS; its effective dipole moment is the sum of the individual dipole moments, thus yielding $\Omega = 2\eta$.

The latter situation has been investigated by Weis *et al.* [11], who measured strongly coupled pairs of Lithium tunneling defects in a KCl matrix. The tunneling defects, which can be well described as TLSs [12,13], interact *via* a dipole-dipole coupling. In the special case investigated by Weis *et al.* this coupling is ferromagnetic ($J > 0$). Due to the strong coupling only one Rabi peak was found. It would be desirable to investigate different pair constellations corresponding to weaker or even antiferromagnetic couplings in order to see the second Rabi frequency.

For $J < 0$, A_1 corresponds to the lower Rabi frequency. Thus, the amplitude of the higher Rabi frequency is suppressed by the temperature dependent factor and we can find a parameter regime, where both amplitudes are of the same order of magnitude. For strong coupling and low temperature, both amplitudes vanish, rendering corrections to the *RWA* of order (η/ω) important. These corrections lead to four new peaks close to the bigger frequency ϵ_1 (while we are in resonance with ϵ_2!) and can be found in Appendix A.2. Yet due to the overall factor of (η/ω) we can not expect to see the new peaks in an experiment.

In the case of resonance with the larger frequency ϵ_1, we get essentially the same results, but the expressions for the two Rabi frequencies and their amplitudes are different and are listed in Appendix A.3. The main difference is the existence of a parameter range in which we get considerable amplitudes for both Rabi frequencies. Furthermore, for $|J| \to \infty$ both Rabi frequencies vanish so that again corrections to the RWA become dominant.

Concerning the third case where both inner frequencies are close to each other, we only considered the situation of resonance with one of them while $(\epsilon_1 - \epsilon_2) \gg \eta$ (see Appendix A.3). The results are very similar to the previously discussed cases, but the corrections are of order $\eta/(\epsilon_1 - \epsilon_2)$ rather than (η/ω).

4 Conclusion

In this paper we have communicated a generalisation of the rotating wave approximation (RWA). First, we propose a formal framework in which for a single spin the RWA appears as the leading order. This formalism allows to easily calculate higher order corrections to the approximation. If, on the other hand, we apply it to a general n-level-system, we can generalise the concept of the RWA by identifying it with the leading order contribution.

Second we have considered a coupled spin pair in an oscillating external field. Although the energetic structure of such a system is well described by two isolated effective spins the time evolution of the system shows some "dynamical entanglement". These effects will disappear in the ferromagnetic strong coupling limit, which makes clear that our results are consistent with previous investigations [2,10,11].

Furthermore our calculations show that there is a situation (concerning spin pairs) where terms beyond the leading order of the RWA become important. This will be the case for an anti-ferromagnetic coupling, where the amplitudes of the leading order are suppressed. We predict that whenever a situation can be modeled by a spin pair with such a coupling, rotary echos should show signals arising from higher order terms in the approximation scheme we proposed. This explicit example shows that, although the RWA seems to be correct for most cases, one can think of situations where the RWA does not suffice in order to separate the 'slow part' from the complete quantum dynamics.

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Appendix A: Calculations and corrections for the pair

A.1 Diagonalization of the pair Hamiltonian

We diagonalize H_{pair} by the transformation

$$\hat{H}_{\text{pair}} = e^{-i(\frac{\alpha}{2}\sigma_y\otimes\sigma_z+\frac{\beta}{2}\sigma_z\otimes\sigma_y)}\, H_{\text{pair}}\, e^{i(\frac{\alpha}{2}\sigma_y\otimes\sigma_z+\frac{\beta}{2}\sigma_z\otimes\sigma_y)}, \tag{A.1}$$

where the angles α and β obey

$$\tan(\alpha+\beta) = -\frac{J}{k} \quad \text{and} \quad \tan(\alpha-\beta) = -\frac{J}{\delta k}\cdot \tag{A.2}$$

This leads to the following values of u in H_{ext}

$$\begin{aligned} u_1 &= \cos(\beta) \quad , \quad u_2 = -\sin(\alpha) \\ u_3 &= \cos(\alpha) \quad , \quad u_4 = -\sin(\beta). \end{aligned} \tag{A.3}$$

A.2 First case: m = 1 and n ≥ 2

In the rotating frame of reference we calculate the effective Hamiltonian following the procedure presented in Section 2.2. Including the dominant corrections of order $|\epsilon_1-\omega|/\omega$, the effective Hamiltonian reads

$$\begin{aligned} \bar{H}_{\text{eff}} = &-\frac{\delta_1}{2}\sigma_z\otimes 1 - \frac{\delta_2}{2}1\otimes\sigma_z + \frac{\eta}{2}(u_1+u_2\sigma_z)\otimes\sigma_y \\ &+\frac{\delta_1}{\omega}\frac{\eta}{(n^2-1)}\sigma_y\otimes(u_3+u_4\sigma_z), \end{aligned} \tag{A.4}$$

where $\delta_1 = \epsilon_1 - n\omega$ and $\delta_2 = \epsilon_2 - m\omega$. Using equation (33), we can calculate the dipole moment up to order η/ω. Considering exact resonance, $\delta_2 = 0$, we obtain

$$\begin{aligned} p^{(1)}(t) = &\frac{1}{4}\{(\frac{\Omega_1}{\eta}A_1 + \frac{\eta}{\omega}B_1)\sin((\omega-\Omega_1)t) \\ &-(\frac{\Omega_1}{\eta}A_1 - \frac{\eta}{\omega}B_1)\sin((\omega+\Omega_1)t)\} \\ &+\frac{1}{4}\{(\frac{\Omega_2}{\eta}A_2 + \frac{\eta}{\omega}B_1)\sin((\omega-\Omega_2)t) \\ &-(\frac{\Omega_2}{\eta}A_2 - \frac{\eta}{\omega}B_1)\sin((\omega+\Omega_2)t)\} \\ &+\frac{\eta}{\omega}B_2\sin(\omega t) \\ &-\frac{\eta}{\omega}B_3\Big\{\sin((\epsilon_1+\eta u_2)t)+\sin((\epsilon_1-\eta u_2)t)\Big\} \\ &-\frac{\eta}{\omega}B_3\Big\{\sin((\epsilon_1+\eta u_1)t)+\sin((\epsilon_1-\eta u_1)t)\Big\} \end{aligned} \tag{A.5}$$

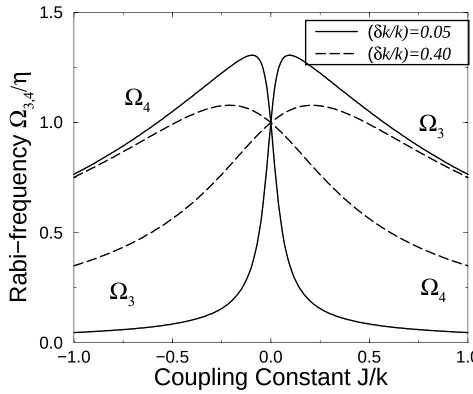

Fig. 3. The two Rabi frequencies $\Omega_{3,4}$ for two different values of $(\delta k/k)$ against (J/k).

with

$$\begin{aligned} B_1 &= \left(\frac{(u_1-u_2)^2}{4} - \frac{n}{n^2-1}u_3u_4\right) r_2(1-r_1) \\ B_2 &= \frac{2n}{n^2-1}(u_3^2+u_4^2)r_1 \\ B_3 &= \frac{1}{n^2-1}u_3(u_3+r_2u_4)r_1, \end{aligned} \tag{A.6}$$

while $A_{1/2}$ and $\Omega_{1/2}$ as well as $r_{1/2}$ were already defined in equation (40).

A.3 Second case: m = 0 and n = 1

All quantities can be obtained from those of the preceding case using the following transformations:

$$n \leftrightarrow m, \quad r_1 \leftrightarrow r_2, \quad u_1 \leftrightarrow u_3 \quad \text{and} \quad u_2 \leftrightarrow u_4. \tag{A.7}$$

As the main results we show the Rabi frequencies plotted *versus* J/k in Figure 3 (corresponding to Fig. 1) and the temperature dependent prefactors A_i plotted *versus* temperature in Figure 4 (corresponding to Fig. 2).

A.4 Third case: n = m = 1

In this case, the effective Hamiltonian reads to lowest order

$$\begin{aligned} \bar{H}_{\text{eff}} = &-\frac{\delta_1}{2}\sigma_z\otimes 1 - \frac{\delta_2}{2}1\otimes\sigma_z + \frac{\eta}{2}\sigma_y\otimes(u_3+u_4\sigma_z) \\ &+\frac{\eta}{2}(u_1+u_2\sigma_z)\otimes\sigma_y, \end{aligned} \tag{A.8}$$

which is very similar to that in equation (A.4). Restricting ourselves to the situation where the energy difference of the two effective TLS's $(\epsilon_1-\epsilon_2)$ is large compared to η, we can use perturbation theory. If the bigger frequency is in

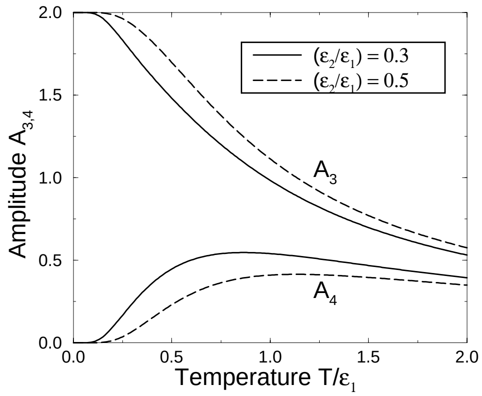

Fig. 4. $r_1(1 \pm r_2)$ for two different values of (ϵ_2/ϵ_1) against (T/ϵ_1)

resonance with the field, $\omega = \epsilon_1$, we obtain for the dipole moment:

$$\begin{aligned} p^{(0)}(t) = &\frac{1}{4}\{(\frac{\Omega_3}{\eta}A_3 + 2\frac{\eta}{\delta_2}B_5)\sin((\omega-\Omega_3)t) \\ &-(\frac{\Omega_3}{\eta}A_3 - 2\frac{\eta}{\delta_2}B_5)\sin((\omega+\Omega_3)t)\} \\ &+\frac{1}{4}\{(\frac{\Omega_4}{\eta}A_4 - 2\frac{\eta}{\delta_2}B_6)\sin((\omega-\Omega_4)t) \\ &-(\frac{\Omega_4}{\eta}A_4 + 2\frac{\eta}{\delta_2}B_6)\sin((\omega+\Omega_2)t)\} \\ &+\frac{\eta}{\delta_2}B_7\sin(\omega t) \\ &-\frac{\eta}{2\delta_2}B_8\Big\{\sin((\bar{\epsilon}_2+\eta u_3)t)+\sin((\bar{\epsilon}_2-\eta u_3)t)\Big\} \\ &-\frac{\eta}{2\delta_2}B_9\Big\{\sin((\bar{\epsilon}_2+\eta u_4)t)+\sin((\bar{\epsilon}_2-\eta u_4)t)\Big\} \end{aligned} \quad \text{(A.9)}$$

with

$$\begin{aligned} &\Omega_{3/4} = \eta(u_3 \pm u_4) \quad A_{3/4} = r_1(1 \pm r_2) \\ &B_{5/6} = r_1 u_1 u_2 (1 \pm r_2) \\ &B_7 = (u_1^2 + u_2^2) r_2 \quad B_{8/9} = r_2 u_{2/1}(u_{1/2} r_1 + u_{2/1}) \\ &\bar{\epsilon}_2 = \epsilon_2 + \frac{\eta^2}{2\delta_2}(u_1^2 + u_2^2). \end{aligned} \quad \text{(A.10)}$$

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