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A Dressed Atom Interpretation of Adiabatic Rapid Passage

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**Title:** A DRESSED ATOM INTERPRETATION OF ADIABATIC RAPID PASSAGE

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**Abstract:** We have considered the problem of adiabatic rapid passage (ARP) in a dressed atom formalism. We find that the conditions for ARP can be understood as conditions for minimizing the probability of dressed atomic-state transitions, and that these probabilities can be used to obtain an estimate of the degree of population reversal. Furthermore, by considering ARP in the dressed atom formalism, the similarity between ARP and the Landau-Zener nonadiabatic transition problem is made quite apparent.
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I. INTRODUCTION

Since the earliest days of nuclear-magnetic resonance (NMR), adiabatic rapid passage (ARP) has been recognized as one of the more important consequences of the transient interaction of radiation and matter (Abragam, 1961; Allen and Eberly, 1975). Basically, the effect of ARP is a population reversal between atomic or molecular states coupled by an electromagnetic field, when either the field or the atomic energy level spacing is rapidly swept through the resonance condition (the resonance condition being defined as $\omega_0 = \omega_f$, with $\omega_0$ being the atomic resonance frequency and $\omega_f$ the frequency of the electromagnetic field). Quite early the conditions for its occurrence were derived by

$$\frac{\omega_1}{T_2} \ll \frac{d}{dt}|\omega_0 - \omega_f| \ll \omega_1^2$$  \hspace{1cm} (1)

where $\omega_1$ and $T_2$ are the Rabi frequency and the transverse relaxation time, respectively; these were given intuitive justification in the now classic discussion of Powles (1958).

Powles' interpretation of the conditions for ARP is based on a Bloch vector model of the interaction process, as illustrated in Fig. 1. In a rotating coordinate system the Bloch vector, describing the state of the atom, precesses with a frequency $\Omega$ about an effective field:

$$\Omega = (\omega_0 - \omega_f)\hat{z} + \omega_1 \hat{\sigma}_z$$

As the system is swept through the resonance condition, the direction of the effective field changes by $\pi$ radians. Thus, if the atom was initially in the state corresponding to $-\hat{z}$ before passage, it will be in the state corresponding to $+\hat{z}$ after passage. The rapidity condition, expressed as the left-hand inequality of Eq. (1), ensures that the rotation of the effective field is fast compared to its relaxation. The adiabaticity condition, expressed as the inequality on the right-hand side of Eq. (1), ensures the Bloch vector's
\[
\Omega = (\omega_0 - \omega_f)^2 + \omega_1 \dot{x}
\]

**Fig. 1.** The Bloch Vector Model of AMP. $\Omega$, $(\omega_0 - \omega_f)$, and $\omega_1$ represent the precessional frequency of the Bloch vector about the effective field, the detuning, and the Rabi frequency, respectively.
ability to follow the change in the effective field's direction. The precessional motion of the Bloch vector must be much faster than the motion of the effective field. The transverse relaxation time is used in Eq. (1), because $T_2 < T_1$; and if population reversal is to occur, the Bloch vector must not be allowed to wander out of the rotating coordinate system $xz$ plane. Powles' description is thus both intuitively clear and simple, and for these reasons has contributed greatly to the understanding of ARP.

In the present report, it is our purpose to discuss the physics of ARP using the dressed atom model of radiative interactions; in particular, we will consider the fully quantized dressed atom, where both the field and the atom are quantized systems (Cohen-Tannoudji, 1977). Our aim is to obtain an interpretation of the conditions for ARP which is as intuitively pleasing as Powles', but one derived from a QED formalism. We will find that in the QED formalism, the conditions for ARP can be interpreted as conditions for minimizing the probability of dressed atomic-state transitions, and that this probability is composed of two parts: a probability for nonadiabatic dressed atomic-state transitions, which is completely analogous to the Landau-Zener inelastic collision probability; and a probability for dressed atomic-state relaxation.

Furthermore, as observed by Lau (1976), there is a similarity between the Landau-Zener inelastic-collision problem and ARP which we will attempt to highlight. In many cases of inelastic atomic or molecular collisions, the process can be viewed intuitively as a transition between adiabatic states near an avoided crossing (Tully, 1976). The adiabatic states are constructed from molecular theory and the Born-Oppenheimer approximation, which presupposes that the electronic motion of the approaching pair can adiabatically adjust itself to changes in the internuclear separation. When the atoms approach each other rapidly, however, this approximation breaks down, and the terms in the Hamiltonian that couple the electronic and nuclear motion give rise to transitions between the adiabatic states. Since the problem is very difficult to handle in the adiabatic representation, Zener worked with the so-called diabatic basis set: a set of basis vectors that minimize the off-diagonal matrix elements of the Hamiltonian (Tully, 1976; Zener, 1932).
was then able to derive an expression for the transition probability between adiabatic states in the vicinity of an avoided crossing. By considering the ARP problem in a dressed atom formalism, it will be possible to both demonstrate the equivalence of the Landau-Zener inelastic-collision problem and ARP, and take full advantage of the already derived result.
II. DRESSED ATOM STATES AS ADIABATIC WAVEFUNCTIONS

It is well known that in the simple case of a two-level atom interacting with a monochromatic field, the Hamiltonian for the field-atom system can be written in terms of a fictitious spin 1/2 and the photon creation/annihilation operators $a$ and $a^\dagger$ (Cohen-Tannoudji, 1977):

$$H = \hbar \left[ \omega_0 S_z + \omega_f a^\dagger a + \lambda S_x (a + a^\dagger) \right]$$

where the zero of energy for the atom is chosen midway between the two states and the zero-point energy of the field has been neglected; $\omega_0$, $\omega_f$, and $\lambda$ represent the energy separation between the two atomic states, the energy of a field photon, and the interaction strength, respectively. Dressed atom states, or adiabatic states in the language of collision processes, are constructed by diagonalizing the Hamiltonian:

$$|u, n\rangle = \cos \frac{\Theta}{2} |e, n\rangle + \sin \frac{\Theta}{2} |g, n + 1\rangle$$

and

$$|d, n\rangle = -\sin \frac{\Theta}{2} |e, n\rangle + \cos \frac{\Theta}{2} |g, n + 1\rangle$$

(Note)

where $|g, n + 1\rangle$ and $|e, n\rangle$ are the unperturbed atom-field states, which correspond to diabatic states, and

$$\tan \Theta = \frac{\lambda \sqrt{n + 1}}{(\omega_0 - \omega_f)} = \frac{\omega_1}{(\omega_0 - \omega_f)}$$

where $(n + 1)$ is the number of photons in the unperturbed field and $\omega_1$ is again defined as the Rabi frequency. The eigenfrequencies corresponding to these dressed atom states are easily found to be

$$\omega_u, n = (n + \frac{1}{2})\omega_f - \frac{1}{2} (\omega_1^2 + \Delta^2)^{1/2}$$

and

$$\omega_d, n = (n + \frac{1}{2})\omega_f + \frac{1}{2} (\omega_1^2 + \Delta^2)^{1/2}$$

and are displayed in Fig. 2 as a function of the detuning $\Delta (\Delta = \omega_0 - \omega_f)$. 

7
Dressed Atom Energy

\[ E_{\text{g},n} \]

\[ E_{\text{g},n+1} \]

\[ E_{\text{g},n-1} \]

\[ \hbar \omega_{\text{u},n} \]

\[ \hbar \omega_{\text{p},n} \]

\[ \hbar \omega_{\text{u},n-1} \]

\[ \hbar \omega_{\text{p},n-1} \]

\[ 0 \]

\[ \hbar \omega_0 \]

\text{PHOTON ENERGY (}\hbar \omega_0\text{)}

**Fig. 2.** Energy Eigenvalues of the Dressed Atom Eigenstates as a Function of Photon Energy. Only two doublets are shown, corresponding to \( n + 1 \) and \( n \) photons in the strong field mode. As described in the text, we can differentiate between two types of relaxation processes for the dressed atom states: intradoublet transitions (indicated by solid lines), and interdoublet transitions (indicated by wavy lines).
In the dressed atom model ARP occurs when the atom-field system enters and leaves the avoided crossing region in the same dressed atom state (Haroche, 1971). For example, if the passage starts with $(\omega_0 - \omega_f) \ll 0$, then the dressed atom wavefunction $|u, n\rangle$ is well approximated by $|g, n + 1\rangle$: The atom is in its lower energy eigenstate and there are $n + 1$ photons in the field. After passage we have $(\omega_0 - \omega_f) \gg 0$ and $|u, n\rangle$ is well approximated by $|e, n\rangle$: The atom is in its upper energy eigenstate. Thus, if the dressed atom state does not change as the system traverses the avoided crossing region, then

$$|g, n + 1\rangle + |e, n\rangle$$

which corresponds to population reversal.

It should be noted that by equating the dressed atom states of Eq. (2) with eigenfunctions of the Hamiltonian we have assumed that $\Delta$ changes slowly in time; this can be regarded as an optical analogue of the Born-Oppenheimer approximation. However, in ARP as in the inelastic-collision problem treated by Landau and Zener, we are interested in the conditions under which this approximation breaks down. We will therefore find it more convenient to consider the problem in the diabatic basis, and by performing the necessary transformation we arrive at the following wave equations:

$$H|g, n + 1\rangle = E_1|g, n + 1\rangle + \frac{\hbar \omega_f}{2} |e, n\rangle$$

$$H|e, n\rangle = \frac{\hbar \omega_f}{2} |g, n + 1\rangle + E_2|e, n\rangle$$

where

$$E_1 = \hbar \left[ (n + \frac{1}{2})\omega_f - \frac{\Delta}{2} \right]$$

$$E_2 = \hbar \left[ (n + \frac{1}{2})\omega_f + \frac{\Delta}{2} \right]$$
If the energy of the field's photons changes linearly in time, then

\[ \frac{1}{\hbar} (E_2 - E_1) = \Delta = \alpha t \]  

(3)

and the problem is formally equivalent to the one solved by Zener (1932). Thus, by approaching the problem of ARP in the framework of dressed atoms, we can immediately make contact with the problem of inelastic collisions treated by Zener, and obtain the probability for an adiabatic passage through the avoided crossing region:

\[ P_{ad} = 1 - \exp\left(-\frac{\omega_1^2}{2|\alpha|}\right) \]

To maximize this probability we obviously want

\[ \omega_1^2 \gg |\alpha| = \frac{d}{dt} |\omega_0 - \omega_\xi| \]

which is the condition expressed by the right-hand side of Eq. (1).
III. RELAXATION OF THE DRESSED ATOM STATE

When the dressed atom states are constructed by quantizing the electromagnetic field, it is well known that for a two-level system a "ladder" of dressed-state doublets results. Cascades down this ladder represent the physical processes by which the field loses photons, and this often takes the form of fluorescence into field modes other than the one strongly coupling the two atomic states (Cohen-Tannoudji, 1977). Thus, we can consider two types of relaxation process, which for generality we will call intra- and interdoublet transitions; these are illustrated in Fig. 2 by the solid and wavy lines, respectively. We should consider the interdoublet transitions as bare atom $T_1$ processes, because in a sense they represent a "spin-lattice" relaxation as defined by Bloembergen, Purcell, and Pound (1948). The spin-lattice interaction describes the process by which energy is transferred from the spin system to a heat reservoir, and in the case of fluorescence the reservoir is composed of all available field modes. Similarly, we should identify the intradoublet transitions as bare atom pure dephasing processes (i.e., $T_2^*$); they result from a change in the phase of the atomic part of the dressed atom wave function, and in the limit that $\Omega \tau_c \ll 1$ (where $\tau_c$ is the time interval over which the intradoublet transition interaction acts), there is no exchange of energy with the fluorescent field modes (Reynaud and Cohen-Tannoudji, 1982). This is not to imply, however, that intradoublet transitions do not require energy. When these transitions result from dephasing collisions, an energy exchange occurs with the kinetic energy of the colliding atoms.

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*a* The bare atom picture corresponds to the diabatic basis set discussed herein. For a comparison between the bare atom and dressed atom descriptions of radiative interactions, the reader is referred to Berman and Salomaa (1982).

*b* It is interesting to note that when this condition is violated ($\omega_1 \tau_c \gg 1$), one would expect the intradoublet transition rate to take on the meaning of the $T_{2e}$ in Redfield's modified Bloch equations (Redfield, 1955).
Recently, Reynaud and Cohen-Tannoudji (1982) considered the problem of dressed atom relaxation in some detail. In particular, they have considered the important special case of fluorescence (with a rate $\Gamma$) as the interdoublet transition mechanism, and atomic dephasing collisions (with a dephasing rate $\gamma$) as the intradoublet transition mechanism. In what follows we draw heavily on their results in developing a simple, approximate means of including in the probability for ARP the effects of these relaxation mechanisms.

It is important to realize that not all the transitions they considered need to be regarded as relaxation transitions in the present problem. In ARP we are only concerned with the probability of emerging from the avoided crossing region of Fig. 2 in a particular unperturbed atomic eigenstate (i.e., $|e, n\rangle$). Thus, if we define $\sigma_{un, un}$ and $\sigma_{2n, 2n}$ as the density matrix elements for the states $|u, n\rangle$ and $|l, r\rangle$, respectively, then the quantity of interest is

$$\Pi_u = \Gamma_{un} \sigma_{un, un}$$

(a similar quantity can be defined as $\Pi_v$). Thus, transitions like $|u, n\rangle \rightarrow |u, n - 1\rangle$ have no effect on $\Pi_u$, and therefore do not behave like relaxation.

We assume that the dressed atom starts the passage in the $|u, n\rangle$ eigenstate with $(\omega_0 - \omega_f) \ll 0$ (i.e., $|u, n\rangle = |g, n + 1\rangle$). For a particular detuning $\Delta$, the evolution of $\Pi_u$ is governed by the equation

$$\frac{d}{dt} \Pi_u(t) = -(2w + \Gamma_{ul} + \Gamma_{lu}) \Pi_u(t) + (w + \Gamma_{ul}) \Pi_u$$

where $\omega$ is the intradoublet transition rate [$\omega = \gamma \omega_1^2/2(\omega_1^2 + \Delta^2)$] and the $\Gamma_{ij}$ are the interdoublet transition rates as illustrated in Fig. 2:

$$\Gamma_{ul} = \Gamma[(\omega_1^2 + \Delta^2)^{1/2} - \Delta^2/4(\omega_1^2 + \Delta^2)]$$
If we now let $\Delta$ be a function of time ($\Delta = |\alpha| t$), but choose $|\alpha|$ small enough so that the passage is adiabatic, then Eq. (4) is a linear differential equation with nonconstant coefficients describing the effects of relaxation on $\Pi_u(t)$ as the field is swept through resonance. The problem of the present section is to compute $\Pi_u(t)$ at some time $\tau$ after the passage: $\tau$ will determine the instant that the field has swept through the "range" of the avoided crossing region, to be defined later, so that $\Pi_u(\tau)$ will approximate the probability for population reversal when relaxation alone is considered.

Integrating Eq. (4) formally with the boundary condition $\Pi_u(-\infty) = 1$, we have for $\Pi_u(\tau)$ the equation

$$\Pi_u(\tau) = 1 + \int_{-\infty}^{\tau} (w + M_u) dt - \int_{-\infty}^{\tau} (2w + M_u + M_\Delta u) \Pi_u(t) dt \quad (5)$$

This equation can be solved to arbitrary precision by the method of successive iterations. However, for the present problem we are primarily concerned with situations for which $\Pi_u(t)$ remains nearly constant throughout the entire passage. Thus, a first approximation is adequate for our purposes, in which case the equation for $\Pi_u(\tau)$ becomes

$$\Pi_u(\tau) = 1 - \int_{-\infty}^{\tau} (w + M_\Delta u) dt$$

Substituting for $w$ and $M_\Delta u$ and performing the integration one obtains

$$\Pi_u(\tau) = 1 - \frac{\omega_1}{2|\alpha|} \left( |\alpha| - \frac{\Gamma}{2} \right) \left[ \frac{\pi}{2} + \tan^{-1} \left( \frac{|\alpha| |\tau|}{\omega_1} \right) \right] - \frac{\Gamma}{2|\alpha|} \left[ |\alpha| |\tau| + (\alpha^2 \tau^2 + \omega_1^2)^{1/2} \right]$$

We now determine the range of the avoided crossing region by defining $\delta$ as the frequency interval about $\omega_0$ over which the doublet separation roughly doubles. We note that this definition is equivalent to
that of Levine and Bernstein (1974), except that they were considering the range of an avoided crossing region between two potential energy curves of a colliding pair of atoms. We thus find that

$$|a| \tau = \omega_1$$

and

$$\Pi_u(\tau) = 1 - \frac{3\omega_1}{\alpha^2} \left( \frac{T_2}{2} + \gamma \right) = 1 - \frac{3\omega_1}{8|a| T_2}$$

(6)

where $T_2$ is the total dephasing time for the bare atom. In order to minimize the probability of relaxation during the passage through the avoided crossing region, we therefore require

$$\omega_1/T_2 >> |a| = \frac{d}{dt} |\omega_0 - \omega_f|$$

which is equivalent to the left-hand side in Eq. (1).

It is interesting to note that a similar expression for $\Pi_u(\tau)$ can be obtained in a less precise, but possibly more intuitive, fashion. We assume that the decay of a dressed atom state can be adequately described in the vicinity of the avoided crossing region by the rate $1/T_2$ (we use the total dephasing rate, so both intra- and interdoublet transitions relax the dressed atom states), so that the probability of finding the dressed atom in any one of the $|u, n\rangle$ eigenstates after a time $\tau$ is

$$\Pi_u(\tau) = \exp(-\tau/T_2)$$

For a passage described by Eq. (3), assigning the width $\delta$ to the avoided crossing region, the probability for a dressed atom to enter and leave the avoided crossing region in one of the $|u, n\rangle$ eigenstates is then

$$\Pi_u(\tau) = \exp\left(-\frac{\omega_1}{|a| T_2}\right) = 1 - \frac{\omega_1}{|a| T_2}$$

for $\omega_1/T_2 << |a|$, which is of the same order of magnitude as Eq. (6).
IV. APPROXIMATE FORMULA FOR THE DEGREE OF POPULATION REVERSAL IN ARP

The preceding discussion has shown that the standard conditions of achieving ARP can be interpreted as conditions for minimizing the probability of dressed atomic-state transitions caused by nonadiabaticity and relaxation. However, it is important to note that the two probabilities $P_{ad}$ and $P_u(\tau)$ were derived under contradictory assumptions regarding the magnitude of the sweep rate $|\alpha|$: In deriving $P_{ad}$ we essentially assumed that the passage was fast enough so that relaxation could be ignored, while in deriving $P_u(\tau)$ we explicitly assumed adiabaticity of the passage. Thus, these two probabilities should be viewed as limiting forms of a more general probability for ARP, $P_{ARP}$, which describes the passage when both nonadiabaticity and relaxation are important.

Unfortunately, a rigorous closed-form expression for $P_{ARP}$ is difficult to obtain; furthermore, if the closed-form expression is too complex, the relevant physics may be obscured. Lau (1976) has analyzed ARP, including relaxation effects, within a semiclassical dressed atom model. He found that as time progresses after the field frequency has swept through resonance, the probability of ARP assumes a particularly simple form. The probability may be separated into two independent factors — one dealing with the adiabaticity of the passage, equivalent to our $P_{ad}$; the other, depending parametrically on the relaxation rates, describing the effects of relaxation. With this result in mind, we assume that $P_{ARP}$ for the fully quantized dressed atom model is amenable to a similar factorization. Specifically, we approximate $P_{ARP}$ by the product of $P_{ad}$ and $P_u(\tau)$:

$$P_{ARP} = (1 - \frac{3|\mu_1|}{8|\alpha|T_2}) [1 - \exp(-\frac{\mu_1^2}{2|\alpha|})]$$  \hspace{1cm} (7)

The value of such an expression will not lie in the high accuracy of its predictions, but in its ability to estimate the physical characteristics of the phenomenon on the "back of an envelope."
As a particular example of the utility of Eq. (7), and in order to get some idea of its accuracy, we consider the case of a two-level atom with density matrix elements $\sigma_{gg}$ and $\sigma_{ee}$ for the lower and upper unperturbed atomic eigenstates, respectively, prior to passage. Since the strong field is considered far from resonance ($\omega_0 - \omega_f << 0$), the dressed atom density matrix elements are given by $\Pi_u = \sigma_{gg}$ and $\Pi_L = \sigma_{ee}$. After the passage through resonance ($\omega_0 - \omega_f >> 0$), the density matrix elements will have been altered, so that

\[ \Pi_u' = \sigma_{ee}' = P_{ARP} \sigma_{gg} + (1 - P_{ARP}) \sigma_{ee} \]  

and

\[ \Pi_L' = \sigma_{gg}' = P_{ARP} \sigma_{ee} + (1 - P_{ARP}) \sigma_{gg} \]  

Obviously, if the density matrix is normalized prior to the passage, it remains normalized after the passage, as can be seen by Eq. (8). Defining the degree of population reversal as the fraction of atoms transferred from the atomic ground state to the atomic excited state, i.e.,

\[ F = \frac{\sigma_{ee}}{\sigma_{gg}} \]

we find that

\[ F = P_{ARP} + (1 - P_{ARP}) \frac{\sigma_{ee}}{\sigma_{gg}} \]  

As discussed by Camparo and Frueholz (1983), the Rabi frequency and relaxation rates are usually determined by experimental conditions and limitations. One is then forced to vary $|a|$ in order to increase the degree of population reversal. The approximate optimum value of $|a|$ can be found by setting the derivative of $P_{ARP}$ with respect to $|a|$ equal to zero. One then finds that for rapid passages
\[ |a|_0 = \frac{\pi \omega_1^2}{2 \ln (1 + \omega_1 T_2 / 3)} \]

for which the probability of ARP becomes

\[ P_{\text{ARP}} = \frac{x - \ln(1 + x)}{(1 + x)} \] (10)

with

\[ x = (\frac{4}{3})\omega_1 T_2 \]

This probability then determines the maximum degree of population reversal, \( F_{\text{max}} \).

In Fig. 3 the dotted line is a plot of \( F_{\text{max}} \) as a function of normalized Rabi frequency \( \omega_1 T_2 \), computed using Eqs. (9) and (10); the initial conditions were chosen so that \( \sigma_{ee}/\sigma_{gg} = 0.27 \). We have also performed numerical calculations of \( F_{\text{max}} \) using a bare atom, two-level density matrix approach (Camparo and Frueholz, 1983). These solutions are displayed as the solid line in Fig. 3. Considering the approximations made in obtaining the closed-form solution, the agreement is rather good; for this particular example the error was \( \sim 10\% \). More important, however, the simple closed-form solution provides a clear intuitive picture of the ARP process.
Fig. 3. The Maximum Degree of Population Reversal as a Function of Normalized Rabi Frequency. The dotted line is a calculation based on the approximate value of $P_{\text{ARP}}$ derived in the text. The solid line is a numerical solution, obtained from a bare atom description of ARP, for the same conditions.
V. SUMMARY

We have shown that a powerful and intuitive approach to adiabatic rapid passage is obtained by using a dressed atom description of the process. Optical analogues of atomic collision processes become quite clear in this description, especially analogues of the Born-Oppenheimer approximation and its violation in the Landau-Zener problem of nonadiabatic transitions. We thus interpret the conditions of ARP as conditions for minimizing the probability of dressed atomic-state transitions during the passage through resonance. Furthermore, we were able to use these probabilities to estimate the degree of population reversal that occurs as a result of ARP, and the agreement with a more accurate treatment of the problem was seen to be quite reasonable.

It is appropriate to mention again that much of the groundwork for this interpretation has been laid by the studies of Reynaud and Cohen-Tannoudji (1982), Horwitz (1975), and Lau (1976). Reynaud and Cohen-Tannoudji have considered the mechanisms of intra- and interdoublet transitions, and Horwitz and Lau have analyzed the nature of adiabaticity in ARP. The present work provides a synthesis of their results as applied to ARP in the fully quantized dressed atom. The strength of the fully quantized dressed atom approach to this problem, rather than the semiclassical dressed atom approach as considered by Lau (1976), is that the distinction between bare atom $T_1$ and pure bare atom $T_2$ relaxation processes is manifestly clear. In our opinion this allows a better intuitive understanding of the way relaxation enters the problem.

Finally, we wish to point out that the similarity of the ARP problem and the collisional problem treated by Zener is not simply a coincidence of the form of certain mathematical equations. Rather, it indicates the underlying importance and generality of the adiabatic theorem of quantum mechanics (Messiah, 1961). Both problems deal specifically with the condition of validity of this theorem, and the consequences of its violation; they simply couch the condition and consequences in terms of different physical variables.
# REFERENCES


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