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Nonadiabatic spin transitions in an inhomogeneous magnetic field

R. D. Hight* and R. T. Robiscoe
Department of Physics, Montana State University, Bozeman, Montana 59715

W. R. Thorson
Department of Chemistry, University of Alberta, Edmonton, Alberta, Canada T6G 2G2
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Model calculations are presented for nonadiabatic passage of an oriented spin through a reversing solenoidal magnetic field; these are useful for comparison with experiments described elsewhere. The model corresponds closely to the well-known Landau-Zener-Stueckelberg model of a two-state curve crossing in molecular collision theory; various modifications appropriate to the Majorana problem are considered. The resulting transition amplitudes are discussed for the $F = 1$ hyperfine component of the $2\,^2S_{1/2}$ metastable state of atomic hydrogen as an example.

I. INTRODUCTION

Nonadiabatic passage of a beam of atoms with oriented spin through an inhomogeneous magnetic field was studied in the 1930's for its potential relevance to measurement of the sign and magnitude of nuclear magnetic moments.[1-10] Early theoretical work by Majorana[4] considered passage of a spin through the neighborhood of a point of zero field and established general concepts of such processes, now commonly called Majorana transitions. Early experiments and their interpretations were limited by experimental techniques then available, but Kellogg, Rabi, and Zacharias[10] were able to determine the sign of the nuclear magnetic moment in the hydrogen atom by this method. Further work of this sort was then overshadowed by the rapid development of beam magnetic resonance methods, which provide a more direct and simpler approach to the nuclear moment properties.

The explicit study of Majorana transitions has since been neglected, and frequently they are mentioned only as something to avoid experimentally because of their possible depolarization of a beam of oriented spins.[11,12] However, as shown experimentally in work presented elsewhere[13,14] a properly designed nonadiabatic passage device can be used to preserve the polarization of an oriented spin while reversing the field direction. This has an important possible application in the case of the $F=1$ hyperfine component of the $2\,^2S_{1/2}$ state of metasable atomic hydrogen (cf. Sec. IV). Calculations presented in this paper were done with this problem in mind.

In this paper we consider Majorana transitions for a model inhomogeneous field which reverses direction; the model field is a reasonable approximation to that actually present in relevant experimental work.[13,14]

According to the theory of angular momentum, transitions among the $2j+1$ magnetic sublevels of a system of fixed angular momentum j, produced by a time-dependent magnetic field $\vec{H}(t)$, are completely characterized by a rotation of the angular momentum vector $\vec{J}$ in three-dimensional (physical) space, and the Eulerian angles prescribing that rotation depend only upon $\vec{H}(t)$ and the effective magnetic moment, not on the value of j. This fact is implicit in Majorana's paper[4] but was more fully developed in later work on the precession of angular momentum in time-dependent fields (see the review by Bloch and Rabi[15]). This means that the $(2j+1)$-dimensional unitary evolution matrix for such a system, $\underline{U}(t, t_0)$, is just the jth irreducible representation matrix for a finite rotation, with Eulerian angles $\alpha(t, t_0)$, $\beta(t, t_0)$, $\gamma(t, t_0)$, and the equations of motion for these angles are j independent. But for the case $j=\frac{1}{2}$ the resulting two-state problem is often familiar in some other physical context. This is the case for our model, which is just the well-known Landau-Zener-Stueckelberg[16-18] model for potential curve crossings in molecular collisions. We can therefore turn the existing analysis of that problem to useful account in the present context of Majorana transitions.

In Sec. II we briefly develop the theory and its application to our model problem. Section III considers possible modifications and improvements to the model, and in Sec. IV we discuss the specific application of the results to the passage of a polarized beam of metastable $(2\,^2S_{1/2})$ hydrogen atoms in the $F=1$ hyperfine state through a reversing solenoidal field.

II. THEORY AND MODEL PROBLEM

Consider a neutral atomic particle of fixed spin j moving classically in a spatially inhomogeneous magnetic field. We assume the field is weak enough that internal vector couplings of the angular momenta forming $\vec{j}$ are unaffected (for example, the hyperfine couplings, if $\vec{j}=\vec{F}=\vec{I}+\vec{J}$), and j is constant. Since the field seen in the rest frame of the particle is time varying, transitions can occur among the magnetic sublevels. Adiabatic passage occurs if the field changes slowly with respect to the Larmor frequency $\omega = g_j\, \mu_B H/\hbar$: then a system in state $|jm\rangle$ of the initial field evolves into a system *in the corresponding state* ($|jm\rangle$) *of the local field* at any later time. If on the other hand the field changes rapidly compared to ω, "diabatic passage" occurs: a system in state $|jm\rangle$ of the initial field remains *in that same (fixed) state,* which is a superposition of many states ($|jm'\rangle$) with respect to subsequent local fields. One then speaks of "nonadiabatic spin flips" or "Majorana transitions." For situations intermediate between adiabatic and diabatic limits, the equations of motion for the system must be solved.

The time-dependent Schrödinger equation for the spin system is $i\hbar(\partial\psi/\partial t)=\mathcal{H}\psi$, with $\mathcal{H}=\vec{\mu}\cdot\vec{H}(t)$, where $\vec{H}(t)$ is the magnetic field in the particle's rest frame and $\vec{\mu}$ is the magnetic moment associated with the spin $\vec{j}$; here we consider atomic systems[19] with

$$\vec{\mu}=g(j)\mu_B\vec{j}\,. \tag{1}$$

For our problem the field is assumed cylindrically symmetrical about the z axis, and the particle moves parallel to this axis at fixed distance r from it (azimuth φ taken to be zero) as shown in Fig. 1. The field can be expressed in terms of its parallel component H_z and perpendicular (radial) component $H_r(=H_x)$, or in terms of the local field magnitude $H(t)$ and direction $\theta(t)$; these depend parametrically on the off-axis distance r (from cylindrical symmetry, $H_r=0$ for $r=0$). We assume that initially the particle is moving in a homogeneous solenoidal field, with H_z constant, $H_r=0$; then it crosses an inhomogeneous region, over which H_z reverses direction and $H_r\neq 0$; finally it emerges into a second (reversed) homogeneous field region. Although the magnetic fields experiences in the remote past and future (beyond the outer ends of the homogeneous field regions) play an important role in any real physical setting, our study here is not directly concerned with their effects; we calculate the time evolution of the system only through this particular field region. Specific prescription of the model field is given later.

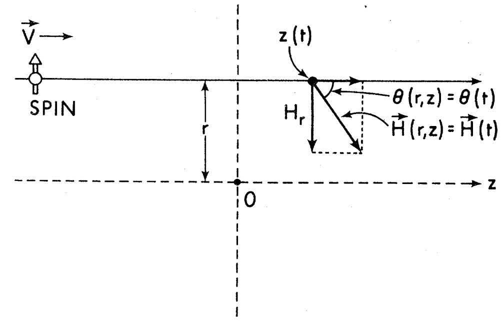

FIG. 1. Particle in a cylindrically symmetric but spatially inhomogeneous magnetic field; the particle moves at constant velocity v, parallel to the cylinder axis (z axis) but at distance r from it.

A. Equations of motion

1. Explicit representation

Explicit representation of the problem is provided by expanding ψ in terms of $(2j+1)$ magnetic sublevel components $|jm\rangle$ defined with respect to some reference axis. If we choose this to be (say) the $+z$ axis, and write

$$\psi=\sum_{m=-j}^{j} A_m(t)|jm\rangle\,, \tag{2}$$

$\underline{A}(t)$, the $(2j+1)$-dimensional representative of ψ, obeys the equation

$$\frac{d\underline{A}}{dt}=-\frac{i}{\hbar}\frac{g}{g_0}\,\omega(t)[\cos\theta(t)\underline{j_z}+\sin\theta(t)\underline{j_x}]\underline{A}(t) \tag{3}$$

where $g=g(j)$, $\omega(t)=g_0\mu_B H(t)/\hbar$ is the local Larmor frequency, and $\underline{j_z}, \underline{j_x}$ are the matrices for the z, x components of angular momentum. It is useful to describe this in interaction picture,

$$A_m(t)=a_m(t)\exp\left(-i\frac{g}{g_0}m\int_0^t \omega(t')\cos\theta(t')dt'\right) \tag{4}$$

yielding the equations

$$\frac{d\underline{a}}{dt}=-i\,(g/g_0)\omega(t)\sin\theta(t)\underline{\underline{K}}\,\underline{a} \tag{5}$$

where the nonzero elements of $\underline{\underline{K}}$ are

$$K_{m\pm 1,m}=\hbar^{-1}\langle jm\pm 1|\hat{j}_x|jm\rangle \times\exp\left(\pm i\frac{g}{g_0}\int_0^t \omega(t')\cos\theta(t')\,dt'\right). \tag{6}$$

Equations (5) are the *equations of motion in diabatic representation.*

It is also useful to describe the motion in terms of basis vectors rotating adiabatically with the field, i.e. with a basis set ($|jm'\rangle$) quantized on the direction, z' of the local field $\vec{H}(t)$. This amounts to a unitary transformation, $\underline{A}(t)=\underline{D}(t)\underline{B}(t)$, where we require that

$$\hbar^{-1}\underline{D}^\dagger(t)[\cos\theta(t)\underline{j}_z+\sin\theta(t)\underline{j}_x]\underline{D}(t)=\underline{K}^A=\hbar^{-1}\underline{j}_{z'} \tag{7}$$

be diagonal at all times. The transformed equations of motion are then

$$\frac{d\underline{B}}{dt}=-i\frac{g}{g_0}\omega(t)\underline{K}^A\underline{B}(t)-\frac{d\underline{D}^\dagger}{dt}\underline{D}\,\underline{B} \tag{8}$$

where

$$K^A_{m'm'}=m',\quad m'=j,j^{-1},\ldots,-j. \tag{9}$$

The matrix $\underline{D}$ achieving this is the $(2j+1)$-dimensional unitary representation of a rotation by $\theta(t)$ in the (x,z) plane,

$$D_{mm'}=d^{(j)}_{mm'}(\theta)=\langle jm|\exp\{i\theta(t)\hat{j}_y/\hbar\}|jm'\rangle. \tag{10}$$

Here $d^{(j)}_{mm'}(\theta)$ is the quantity so defined by Edmonds[20] and $\hat{j}_y$ the y component of the angular momentum operator. From (10) we obtain

$$-\frac{d\underline{D}^\dagger}{dt}\underline{D}=-\frac{i}{2}\frac{d\theta}{dt}\underline{j}_y; \tag{11}$$

again transforming to interaction picture, we put

$$B_{m'}(t)=b_{m'}(t)\exp\left(-i\frac{g}{g_0}m'\int_0^t\omega(t')dt'\right) \tag{12}$$

and obtain the *equations of motion in adiabatic representation*,

$$\frac{d\underline{b}}{dt}=\frac{1}{2}\frac{d\theta}{dt}\underline{P}\,\underline{b}, \tag{13}$$

where the nonzero elements of P are

$$P_{m'\pm1,m'}=-i\langle jm'\pm1|\hat{j}_y|jm'\rangle\exp\left(\pm i\frac{g}{g_0}\int_0^t\omega(t')dt'\right). \tag{14}$$

2. *Unitary time-development matrices*

Unitary time-development matrices can be defined for either representation,

$$\underline{a}(t)=\underline{U}^D(t,t_0)\underline{a}(t_0), \tag{15a}$$

$$\underline{b}(t)=\underline{U}^A(t,t_0)\underline{b}(t_0). \tag{15b}$$

$\underline{U}^D$ and $\underline{U}^A$ obey Eqs. (5) and (13), respectively, with initial conditions

$$\underline{U}^D(t_0,t_0)=\underline{1}=\underline{U}^A(t_0,t_0). \tag{15c}$$

We assume the system is in a known state at a time t_-, somewhere in the initial homogeneous field region, and we wish to know its state at time t_+, somewhere in the final homogeneous field region; we need to compute the interaction picture S matrices

$$\underline{S}^D=\underline{U}^D(t_+,t_-),\quad \underline{S}^A=\underline{U}^A(t_+,t_-). \tag{16}$$

According to angular momentum theory the system's behavior always corresponds to a physical rotation of the vector $\vec{J}$ in three-dimensional space; hence the matrices $\underline{U}^D$, U^A are just $(2j+1)$-dimensional representations of finite rotation (cf. Edmonds[20]) and are fully determined by three Euler angles α,β,γ. The forms for $j=1$ and $j=\frac{1}{2}$, which are of explicit interest to us, are

$j=1$:

$$U_{11}=[U_{\bar{1}\bar{1}}]^*=\tfrac{1}{2}[1+\cos\beta]\,e^{i(\alpha+\gamma)}, \tag{17a}$$

$$U_{10}=-[U_{\bar{1}0}]^*=2^{-1/2}\sin\beta\,e^{i\alpha}, \tag{17b}$$

$$U_{01}=-[U_{0\bar{1}}]^*=-2^{-1/2}\sin\beta\,e^{i\gamma}, \tag{17c}$$

$$U_{1\bar{1}}=[U_{\bar{1}1}]^*=\tfrac{1}{2}[1-\cos\beta]\,e^{i(\alpha-\gamma)}; \tag{17d}$$

$j=\frac{1}{2}$: $(+=\frac{1}{2},\ -=-\frac{1}{2})$

$$U_{++}=U^*_{--}=\cos(\beta/2)e^{i(\alpha+\gamma)/2}, \tag{18a}$$

$$U_{+-}=-U^*_{-+}=\sin(\beta/2)e^{i(\alpha-\gamma)/2}. \tag{18b}$$

Appropriate superscripts A or D should be attached to the Eulerian angles α,β,γ. The equations of motion for these angles, and their solutions, are j independent. This permits us to solve our model problem for all j, including the case of explicit interest $j=1$, by determining the Euler angles from the solutions for $j=\frac{1}{2}$, which are those of a familiar two-state problem in molecular collision theory.

3. *Symmetry properties*

In addition to cylindrical symmetry the reversing model field considered here has certain symmetries about its midpoint $z=0$ $(t=0)$:

$$H_z(-z)=-H_z(z),\quad H_r(-z)=H_r(z). \tag{19}$$

If we take $t_-=-t_+$, the unitary development matrices have corresponding symmetries (for brevity, proof is omitted):

$$U^D_{mm'}(-t,0)=[U^D_{-m,-m'}(t,0)]^*,$$
$$U^A_{mm'}(-t,0)=[U^A_{-m,-m'}(t,0)]. \tag{20}$$

Since $\underline{S}=\underline{U}(t_+,-t_+)=\underline{U}(t_+,0)\underline{U}(0,t_+)$, additional simplifications result; in terms of the Eulerian angles,

$$\gamma^D(t_+,-t_+)=-\alpha^D(t_+,-t_+),$$
$$\gamma^A(t_+,-t_+)=\alpha^A(t_+,-t_+), \tag{21}$$

and for both adiabatic and diabatic angles

$$\beta(t_+,-t_+)=2\beta(t_+,0),\quad \alpha(t_+,-t_+)=\alpha(t_+,0). \tag{22}$$

B. Model problem solution

The model field assumed is as follows (see Fig. 2):

$$H_z = +H_0, \quad H_r = 0, \quad z \geq z_0; \tag{23a}$$

$$H_z = -H_0, \quad H_r = 0, \quad z \leq -z_0; \tag{23b}$$

$$H_z = H_0(z/z_0), \quad -z_0 \leq z \leq z_0; \tag{23c}$$

in the middle region, H_r is fixed by $\operatorname{div}\vec{H} = 0$,

$$H_r = -(r/2z_0)H_0 = -\rho H_0, \quad -z_0 \leq z \leq z_0. \tag{23d}$$

With fields defined by (23), the angle θ $[\tan^{-1}(H_r/H_z)]$ goes from $-\pi$ to 0. In the inhomogeneous region,

$$(g/g_0)\omega(t)\sin\theta(t) = g\mu_B H_r/\hbar = -(g\mu_B H_0 \rho/\hbar), \tag{24a}$$

$$(g/g_0)\omega(t)\cos\theta(t) = g\mu_B H_z/\hbar = (g\mu_B H_0\rho/\hbar)(vt/\rho z_0), \tag{24b}$$

where v is the particle speed. It is convenient to define[21]

$$s = (g\mu_B H_0 \rho/\hbar)t \tag{25a}$$

and the characteristic variable $\tau(s)$ given by

$$\tau(s) \equiv \cot\theta(s) = -(v/\rho z_0)t = -A^{-1}s, \tag{25b}$$

where the fundamental parameter A is given by

$$A = (g\mu_B H_0 z_0/\hbar v)\rho^2 \equiv \alpha_0 \rho^2. \tag{25c}$$

Equations (5) can then be compactly written with s as independent variable,[21]

$$d\underline{a}/ds = -i\underline{K}(s)\underline{a}(s) \tag{26a}$$

where $\underline{K}(s)$ is given by Eq. (6) with exponent

$$\eta = \frac{g}{g_0}\int_0^t \omega(t')\cos\theta(t')\,dt' = \int_0^s \tau(s')\,ds'; \tag{26b}$$

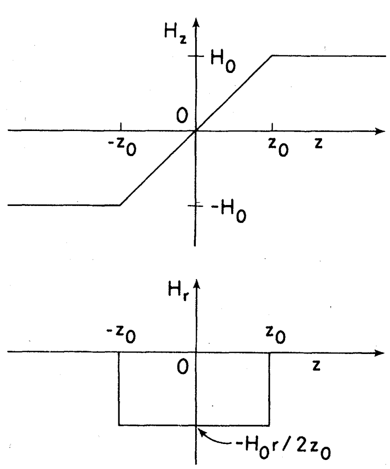

FIG. 2. Model inhomogeneous magnetic field.

for our specific model,

$$\eta(s) = -s^2/2A. \tag{26c}$$

The equations of motion in adiabatic representation (13) can also be expressed in terms of $\tau(s)$,[21] or they can be expressed in terms of θ as independent variable and a characteristic function called the adiabaticity,[22]

$$\alpha(\theta) = \frac{\omega(t)}{d\theta/dt}; \tag{27a}$$

the exponent in Eqs. (13) is the adiabatic phase

$$\lambda = \frac{g}{g_0}\int_0^t \omega(t')\,dt' = \int_{-\pi/2}^{\theta} \alpha(\theta')\,d\theta' \tag{27b}$$

$$= \int_0^s [1 + \tau^2(s')]^{1/2} ds'. \tag{27c}$$

Using these parametric descriptions, it is evident that for $j = \frac{1}{2}$ our model problem is identical[22] to the well known Landau-Zener-Stueckelberg[16-18] model of a two-state curve crossing, which assumes a constant coupling matrix element (here due to the transverse field H_r) and a linear diagonal element difference (here, the Zeeman splitting due to the parallel field H_z)—more generally, $\tau(s)$ is linear in s.[21] Discussions and modifications of the theory of inelastic coupling in the LZS problem[16-18,21,23,24] are therefore directly applicable here.

For the moment let us assume the particle enters the inhomogeneous field region at $t_-(=-t_+)$ and leaves it at $t_+(z_0 = vt_+)$. We wish to compute the state vector just after exit, $\underline{A}(t_+ + \epsilon)$, from its value just before entry, $\underline{A}(-t_+ - \epsilon)$ (in the limit as $\epsilon \to 0^+$). Using Eq. (4) we find

$$A_m(t_+) = \sum_{m'} \exp[-i(m-m')\eta(t_+)]\, S^D_{mm'} A_{m'}(-t_+); \tag{28a}$$

via the relation $\underline{B}(t) = \underline{D}^\dagger(t)\underline{A}(t)$ and Eq. (12) we can also express this as

$$A_m(t_+) = \sum_{m'} \exp[-i(m-m')\lambda(t_+)] \times S^A_{m,-m} D^\dagger_{-m',m'}(-t_+ - \epsilon) A_{m'}(-t_+ - \epsilon). \tag{28b}$$

[Note that $\underline{D}(t_+ + \epsilon) = \underline{1}$, while $\underline{D}(-t_+ - \epsilon)$ represents a rotation by $\theta = -\pi$.] As will be seen below, Eq. (28b) is often more convenient to express results.

Since the inhomogeneous region is finite $(z_0 \geq z \geq -z_0)$, the LZS model and the corresponding equations of motion are really valid only over a finite domain of the independent variable $(s_+ \geq s \geq -s_+; -\pi + \theta_+ \leq \theta \leq -\theta_+, \; \theta_+ = \tan^{-1}\rho)$. However, at least for reasonable values of the off-axis parameter ρ, velocity v, and field strength H_0, it will turn out that nonadiabatic transitions occur in a localized region around $z\approx 0$, well inside the boundaries of the inhomogeneous region. This means that the matrix elements of $\underline{S}^D$ or $\underline{S}^A$ can be evaluated (except for phase in the case of $\underline{S}^D$) by taking the limit $t_+\to\infty$. This hypothesis corresponds exactly to the analogous treatment of the LZS model problem for curve crossings[16-18] and has the same general conditions of validity.[21,23] In Sec. III we examine a modified model taking the finite cut-offs into account.

It can be shown that in the limit $t_+\to\infty$ the parameters of $\underline{S}^A$ all remain finite; this isn't true of $\underline{S}^D$. The results are those of the famous LZS formula for $j=\frac{1}{2}$, which yields the following results for the Eulerian angles

$\beta^A(+\infty,-\infty)$ and $\alpha^A(+\infty,-\infty)$:

$$\sin^2[\beta^A(+\infty,-\infty)/2]=\exp[-\pi A/2]\,, \tag{29a}$$

$$\alpha^A(+\infty,-\infty)=\operatorname{Arg}\Gamma(iA/4)+(A/4)-(A/4)\ln(A/4)+\pi/4. \tag{29b}$$

From Eqs. (29) we may compute the matrix elements for any j value required. Figure 3 depicts the moduli of elements of $\underline{S}^A$ as a function of the parameter A, for $j=1$, and Fig. 4 shows the angle $\phi=\alpha^A(+\infty,-\infty)$ vs A.

For the evaluation of $\lambda(t_+)$, which appears in Eq. (28b), extension of the limit $t_+\to\infty$ is inappropriate, since (27b) then diverges. Instead, we calculate the integral to the finite cut-off point [or, alternatively, use an even more realistic model of the field to evaluate $\lambda(t_+)$]. Employing our simple model with the cutoff at $\pm z_0$, we obtain

$$\lambda(t_+)=\tfrac{1}{2}\alpha_0\{(1+\rho^2)^{1/2}-\rho^2\ln\rho+\rho^2\ln[1+(1+\rho^2)^{1/2}]\}. \tag{30}$$

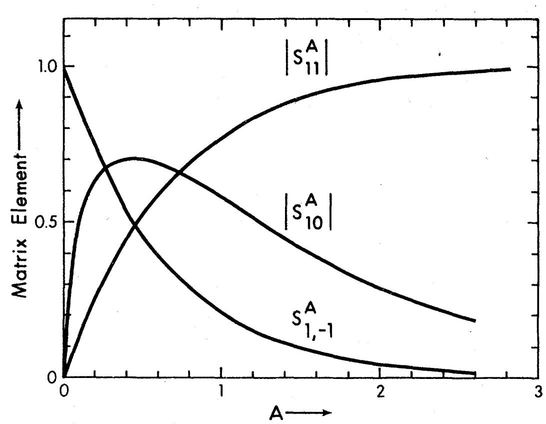

FIG. 3. Moduli of the matrix elements S^A_{11}, S^A_{10}, and $S^A_{1\bar{1}}$ vs the parameter $A=\alpha_0\rho^2$.

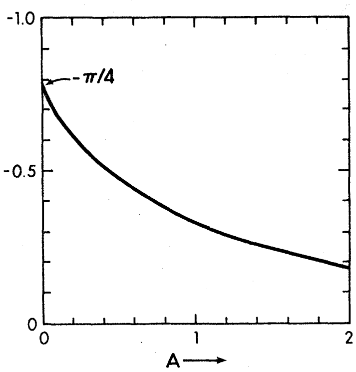

FIG. 4. Phase $\phi=\alpha^A(+\infty,-\infty)$ vs the parameter A. Note that this only changes by $\pi/4$ over the entire range.

III. MODIFICATIONS

The solution given in Sec. II assumes a stylized model field [Eqs. (23)], with discontinuous cut-offs at $\pm z_0$, and it assumes that $\underline{S}^A$ can be accurately computed taking $t_+\to\infty$ (which yields the LZS result). In reality, the coupling region does have a finite width and it is smoothly rather than discontinuously connected with the adjoining homogeneous field regions. In this section we examine two modifications of the theory which take these assumptions into account and allow us to form some estimate of their effects. The first treatment explicitly considers the fact that the inhomogeneous region is finite; the second is a modified model which rounds off the discontinuities at $\pm z_0$.

A. Finite cut-off model

The LZS model equations [Eqs. (26a) and (26c)] have known analytical solutions for $j=\frac{1}{2}$ (parabolic cylinder functions). Therefore if we evaluate the solutions at the finite arguments $\pm s_+$, we can evaluate $\underline{S}^A=\underline{U}^A(t_+,-t_+)$ exactly. This approach was originally suggested by Heinrichs[24] for the potential curve crossing problem, and though it has been shown to be inappropriate for that context, it is quite applicable here.

The dimensionless argument of the parabolic cylinder functions in this problem has modulus $(s/A^{1/2})$, which is equal to $\alpha_0^{1/2}$ at s_+ [Eq. (25c)]. For small α_0 (high velocity, very weak field), the extrapolation $s_+\to\infty$ is obviously inappropriate. However, the LZS result does correctly predict the diabatic behavior of the system in the diabatic passage limit, and is incorrect only in its estimate of the small deviations from that behavior for small α_0. In the applications of interest to us (cf. Sec. IV), these errors are unimportant, the region of greater interest being that with $\alpha_0 \gtrsim 1$. Should accurate results be required for $\alpha_0 \ll 1$, explicit evaluation of the solutions (as confluent hypergeometric series in α_0) can be performed; the leading terms can also be obtained using the perturbation expansion of the usual integral equation for $\underline{U}^D$. However, this sort of application of the finite cut-off model does not concern us here.

A more relevant application is to the case where α_0 is moderately large (though finite), and we are concerned with the effects of variation in the off-axis parameter ρ, which defines the angle θ at cut-off, $\tan^{-1}\theta(S_+) = -\rho$ (we consider $\rho \lesssim 0.5$; in experimental applications, $\rho \lesssim 0.25$). Under these circumstances, we find the following results [we give $\underline{S}^A = \underline{U}^A(t_+, -t_+)$ for $j = \frac{1}{2}$; Eqs. (18) then determine the Eulerian angles β^A, α^A]:

$$[\underline{S}^A]_{+-} = \{e^{-\pi A/4}[\,|\overline{D}_0|^2 - (\rho^2/4)|\overline{D}_{\overline{1}}|^2] + \rho(1 - e^{-\pi A/2})^{1/2}\,\mathrm{Re}[(\overline{D}_0\overline{D}_{\overline{1}})^*\exp(i\Phi_D)]\}, \tag{31a}$$

$$[\underline{S}^A]_{++} = e^{i\Phi_A}\{(1 - e^{-\pi A/2})^{1/2}[(\overline{D}_0^*)^2 - (\rho^2/4)(\overline{D}_{\overline{1}})^2\exp(-2i\Phi_D)] - \rho e^{-\pi A/4}[(\overline{D}_0^*\overline{D}_{\overline{1}})e^{-i\Phi_D}]\}, \tag{31b}$$

where the phases Φ_D, Φ_A are given by

$$\Phi_D = (\alpha_0/2) + (A/4)\ln\alpha_0 - \mathrm{Arg}\,\Gamma(iA/4) - \pi/4 \tag{31c}$$

and

$$\Phi_A = -\mathrm{Arg}\,\Gamma(iA/4) + (A/4)\ln(A/4) - \pi/4 - (\alpha_0/2)[(1+\rho^2)^{1/2} - 1] - (A/2)\ln\{[1 + (1+\rho^2)^{1/2}]/2\}; \tag{31d}$$

the quantities $\overline{D}_0$ and $\overline{D}_{\overline{1}}$ are closely related to the relevant parabolic cylinder functions and can be well represented by asymptotic expansions for moderate to large α_0. Over the range of α_0 and ρ of interest to us, they differ from unity by less than 5%. Equations (31) then give us some idea of the deviations from the LZS formula, which are of two sorts: (1) secular deviations proportional to ρ^2, (2) oscillatory deviations mainly proportional to ρ. The latter are transients caused by the discontinuities in the fields at $\pm z_0$, and will be damped out if these are smoothed out as is the case in the real system.

B. Generalization of model using Stueckelberg's method

Some parameters of S^A can be calculated for more general models than that of Eqs. (23), using a partly analytical method originally developed by Stueckelberg[18] to discuss the LZS model problem. Stueckelberg obtained a formula which gives the modulus of the transition amplitude in terms of a certain integral in the complex s plane. The conditions for validity of this formula have been discussed by several authors.[21,23,25] The Stueckelberg formula is[18,23,25]

$$\sin[\beta^A(+\infty, -\infty)/2] = \exp[\mathrm{Re}(i\delta)] \tag{32a}$$

where δ is a contour integral defined as follows: let $\tau(s)$ be the function defined earlier [Eq. (25b)]; then

$$\delta \int_{0+}^{+i} (1+\tau^2)^{1/2}(d\tau/ds)^{-1}\,d\tau \tag{32b}$$

where the integral is on the pure imaginary τ axis. If $\tau(s)$ is given by the linear form in Eq. (25b), one obtains $\delta = iA\pi/4$ and the LZS result. However, we can use more general forms for $\tau(s)$, provided the integral (32b) can be evaluated using the appropriate analytic continuations.

In our problem the dependency relation between H_r and H_z makes this simpler. Let us write

$$\overline{z} = z/z_0, \quad H_z = H_0 f(\overline{z});$$

then

$$H_r = -\rho H_0 f'(\overline{z}), \tag{33}$$

and we have

$$s = \frac{g}{g_0}\int_0^t \omega(t')\sin\theta/(t')\,dt' = \alpha_0\rho f(\overline{z}), \tag{34a}$$

from which, given the definition of $\tau(s)$, it follows that

$$\delta = A\int_0^{\overline{z}(i)} \{[f'(\overline{z})]^2 + [f(\overline{z})/\rho]^2\}^{1/2} d\overline{z}/\rho, \tag{34b}$$

where $\tau(\overline{z}(+i)) = +i$.

As a suitable modification, consider the form $f(\overline{z}) = \tanh\overline{z}$. This function has the same slope at $z = 0$ as the LZS model, and the same asymptotic limits of ± 1 in the homogeneous regions as does Eqs. (23), but instead of sharp cut-offs at $\pm z_0$ the inhomogeneous region is tapered off gradually around $\pm z_0$("rounding" of the cutoffs is probably overemphasized in this model). We find

$$\delta = i(A\pi/4)F(\rho) \tag{35a}$$

where

$$F(\rho) = \frac{8}{\pi}\int_0^{\pi/2} \frac{\cos^2 y\,dy}{(1 - 4\rho^2\sin^2 y)^{1/2}[1 + (1 - 4\rho^2\sin^2 y)]^{1/2}} \tag{35b}$$

Figure 5 depicts $F(\rho)$ as a function of ρ; $F(0)=1$, while at the upper limit $\rho=\frac{1}{2}$, $F(\frac{1}{2})=4-(8/\pi)=1.453\,52$. For the range of ρ values of most interest to us, the value of F differs by at most 5% from the LZS value $F=1$. This tells us that the LZS prediction for the modulus angle $\beta^A(t_+,-t_+)$ is not grossly incorrect, and indicates the errors likely from defects of the model assumed in Eqs. (23). If we were to use Stueckelberg's formula to compute transition probabilities for this generalized model, (1) we should compute $\lambda(t_+)$ differently for use in Eq. (28b), and (2) the inelastic phase $\alpha^A(+\infty,-\infty)$ cannot be computed by Stueckelberg's method, though we may guess it does not differ much from the LZS value (29b).

IV. APPLICATIONS

A. General considerations

For any experimental applications, we must keep in mind that the above results give the evolution of the system only over the inhomogeneous field region (from $-t_+$ to t_+); the state of the system was assumed to be fully specified at $t_-=-t_+$. Secondly, an experiment involves a beam of particles of finite width and resolution, i.e. an average over a distribution of the parameters ρ, v, etc. is required.

First consider the embedding of the evolution predicted by Eqs. (28) in a real physical situation. In general, the system state at time t_- depends on its evolution from some "state preparation" which prescribes the state at some still earlier time t_0, perhaps in an entirely different apparatus. Furthermore, the result of the experiment will normally be monitored by measurements at some time t_f still later than t_+. Detailed discussion of system evolution from t_0 to t_- and from t_+ to t_f is obviously beyond the scope of this paper, but it may be reasonable to assume that the evolution

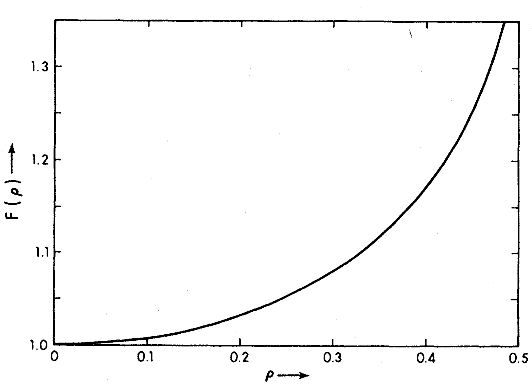

FIG. 5. Modifying exponent factor $F(\rho)$ as a function of ρ [cf. Eqs. (35)].

over these intervals is simple (to within acceptable error limits). One might, for instance, assume that the system develops adiabatically, which is the presumed case in the experimental work to which this study has direct application.[13,14,25] But measurables can be directly affected by these external regions, even in such simple cases, if interference effects due to phase coherence are observed. For example, in our earlier discussion of the model field (Sec. IIB) we assumed that immediately before and after the inhomogeneous field region, the particle passes through regions of homogeneous solenoidal fields, $\mp H_0$ in the z direction. Even though no transitions occur in such homogeneous fields, the relative phases of (stationary state) components making up a coherent state of the system are altered. In principle, these phase changes can be predicted, given a map of the fields, the trajectory and velocity, etc.

In this problem the particle beam (assumed concentric with the solenoid axis) has a finite width, with significant density for off-axis parameters $\rho \gtrsim 0.25$, and for comparison with experiment one must average over this distribution.

B. Specific example

The experimental problem of direct interest to us involves hyperfine components of the metastable $2^2S_{1/2}$ level of atomic hydrogen. Figure 6 shows the schematic behavior of energy levels vs magnetic field for this system. In strong fields (where the electronic Larmor frequency greatly exceeds the hyperfine splitting) a beam can be prepared with only the "α" electron spin levels populated.[25] If this beam then passes *adiabatically* from the strong-field region into the weak field region considered here, the $m=+1, 0$ sublevels of the $F=1$ hyperfine component are the states populated. Nonadiabatic reversal of the *weak* field, as considered here, can then be used to selectively populate the $F=1$, $m=-1$ sublevel; this has been done experimentally by Robiscoe.[25] In part this paper is an attempt to explain the observed $m=-1$ population as function of the magnetic field H_0 characterizing the weak field region.

The most significant feature of this population is its oscillatory behavior vs field. According to the theory this is the result of a coherent population of *two adiabatic* sublevels ($m'=+1, 0$) at the "entry time" t_-. Note that since the *diabatic states* are here defined with respect to the final (reversed) magnetic field, this corresponds to coherent initial *diabatic* populations of $m=-1, 0$. We are then interested in the probability of occupation of the diabatic sublevel $m=-1$, which corresponds also to the *adiabatic* sublevel $m' = -1$, at "exit time" t_+. We assume that at $t = t_-$, the *diabatic* amplitudes are

$$|A_{-1}(t_-)| = |A_0(t_-)| = 1/\sqrt{2}\ , \qquad (36a)$$

$$A_1(t_-) = 0, \qquad (36b)$$

$$\mathrm{Arg}[A_{-1}(t_-)] - \mathrm{Arg}[A_0(t_-)] = \xi . \qquad (36c)$$

The phase ξ has a value which in general depends on the field H_0, the velocity of passage, and also on parameters characterizing the system's earlier evolution through the strong field region. Then from Eqs. (28) the probabilities $|A_m(t_+)|^2$ are

$$|A_1(t_+)|^2 = \tfrac{1}{2}(1-e^{-\pi A/2})\{(1+e^{-\pi A/2}) - 2\sqrt{2}\,e^{-\pi A/4}(1-e^{-\pi A/2})^{1/2}\cos(\lambda_+ - \phi - \xi)\}, \qquad (37a)$$

$$|A_0(t_+)|^2 = \tfrac{1}{2}\{[(1-e^{-\pi A/2})^2 + e^{-\pi A}] + 2\sqrt{2}\,e^{-\pi A/4}(1-e^{-\pi A/2})^{1/2}\cos(\lambda_+ - \phi - \xi)\}, \qquad (37b)$$

$$|A_{-1}(t_+)|^2 = \tfrac{1}{2}e^{-\pi A/2}\{(2-e^{-\pi A/2}) + 2\sqrt{2}\,e^{-\pi A/4}(1-e^{-\pi A/2})^{1/2}\cos(\lambda_+ - \phi - \xi)\}, \qquad (37c)$$

where $\lambda_+ \equiv \lambda(t_+)$ and $\phi \equiv \alpha^A(+\infty, -\infty)$ are given respectively by Eqs. (30) and (29b).

$|A_{-1}(t_+)|^2$ is the probability of occupying the state $m = -1$ after reversal of the weak field H_0; for small A (weak coupling, small H_0, high velocity) diabatic passage occurs, i.e. the sudden approximation is valid and the spin is "flipped" with respect to the suddenly reversed field, so that $|A_{-1}(t_+)|$ remains $1/\sqrt{2}$, as before field reversal. For large A, on the other hand, adiabatic passage occurs and $|A_{-1}(t_+)|^2$ goes to zero, since the spin then follows the reversing field.

In Fig. 7 we have depicted the probability of a nonadiabatic "spin flip," $|A_{-1}(t_+)|^2$, for some representative choices of physical parameters. In order to emphasize the effect of field regions outside the inhomogeneous region considered explicitly here, we have calculated the phase ξ for a schematic model: we will assume that prior to entry of the inhomogeneous region at $t = t_-$, the particle beam travels through a *homogeneous z* field $-H_0$, for a distance $d = v(t_- - t_0)$ (it enters this homogeneous region at $t = t_0$); prior to t_0, the particle has travelled through a strong-field region whose properties are independent of H_0, and we can then assume that at t_0 the relative phase of $A_{-1}(t_0)$ and $A_0(t_0)$ is a constant ξ_0, *whose value may depend on v,* but not upon H_0. Then it is easy to show that

$$\xi = \xi_0 - \alpha_0 \xi_1$$

where α_0 was defined in Eq. (25c), and $\xi_1 = d/z_0$. In Figure 7 we have (arbitrarily) taken $\xi_0 = -\pi/2$ and $d/z_0 = 1$. With off-axis parameter $\rho = 0.2$, we have $\lambda_+ = 0.5562\alpha_0$ from Eq. (30), while $\xi = \xi_0 - \alpha_0$ and $\phi \leq \pi/4$. The obvious conclusion is that while the *envelope* of the curve for $|A_{-1}(t_+)|^2$ is determined

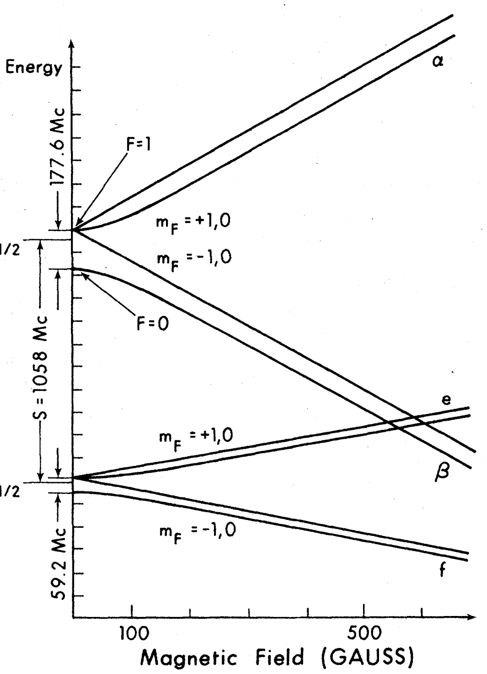

FIG. 6. Schematic diagram of the energy levels of the $2\,^2S_{1/2}$ state of atomic hydrogen, vs magnetic field H.

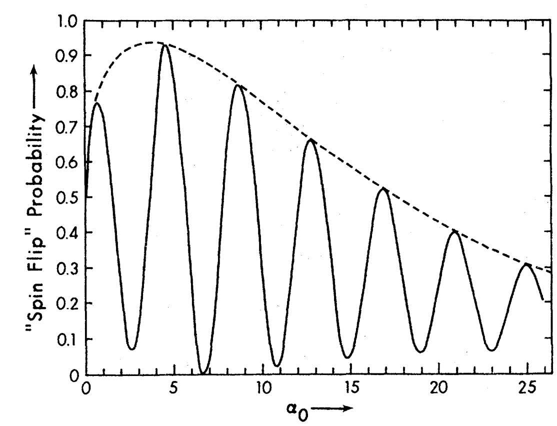

FIG. 7. "Spin-Flip" probability $|A_{-1}(t_+)|^2$ vs α_0 (i.e., vs magnetic field for fixed v, ρ) for off-axis parameter $\rho = 0.2$. The range of α_0 shown corresponds to $H_0 \leq 3$ G for $v = 1.0 \times 10^6$ cm sec^{-1} and $z_0 = 1$ cm. The "external phase" $\xi = -(\pi/2 + \alpha_0)$ (see text). The dashed curve shows the envelope of maxima.

by the coupling region parameters, this is not the case for the frequency and absolute phase of the coherent oscillations, which are likely to be dominated by the "external region" characteristics, even though these depend on the field strength or α_0 in the same way as λ_+ does. In the present example, even the modest ratio $d/z_0=1$ triples the frequency of oscillations.

The primary interest in this problem experimentally lies in the possibility of the preparation of a beam with exclusive populations of the sublevels $m=-1, 0$ of the $F=1$ hyperfine component by sudden field reversal; if the system can then be adiabatically returned to strong field, the $m=0$ component can readily be defocused, leaving an atomic system of unique polarization.

The immediate conclusions to be drawn from this simple theoretical study are that (1) the probability envelope for the Majorana transitions occurring on passage through a reversing field system of this type can certainly be understood by adequate model study, but we should emphasize that (2) the frequency and phase of the coherent oscillations observed cannot be predicted without taking into account the field characteristics external to the field-reversing region.

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*Present address: Dept. of Physics and Astronomy, University of Toledo, Toledo, Ohio 43606.

[1]J. M. B. Kellogg, I. I. Rabi, and J. R. Zacharias, Phys. Rev. 50, 472 (1936).

[2]H. C. Torrey and I. I. Rabi, Phys. Rev. 51, A379 (1937).

[3]S. Millman and J. R. Zacharias, Phys. Rev. 51, A380 (1937).

[4]E. Majorana, Nuovo Cimento 9, 43 (1932).

[5]T. E. Phipps and O. Stern, Z. Phys. 73, 185 (1931).

[6]R. Frisch and E. Segre, Z. Phys. 80, 610 (1933).

[7]I. I. Rabi, Phys. Rev. 49, 324 (1936).

[8]J. Schwinger, Phys. Rev. 51, 648 (1937).

[9]L. Motz and M. E. Rose, Phys. Rev. 50, 348 (1936).

[10]J. M. B. Kellogg, I. I. Rabi, and J. R. Zacharias, Phys. Rev. 49, A641 (1936).

[11]N. F. Ramsey, *Molecular Beams*, 1st ed. (Oxford U.P., 1963), p. 401.

[12]C. W. Drake, Jr., in *Methods of Experimental Physics*, edited by L. Marton (Academic, New York, 1967), Vol. 4B, p. 250.

[13]R. D. Hight, Ph.D. thesis (Montana State University, 1975) (unpublished); and unpublished work.

[14]R. D. Hight, *Abstracts of Papers for the Ninth International Conference on Physics of Electronic and Atomic Collisions*, edited by J. S. Risley and R. Geballe (Univ. of Washington Press, Seattle, 1975).

[15]F. Bloch and I. I. Rabi, Rev. Mod. Phys. 17, 237 (1945).

[16]L. Landau, Phys. Z. Sowjetunion 2, 46 (1932).

[17]C. Zener, Proc. Roy. Soc. (London) A137, 696 (1932).

[18]E. C. G. Stueckelberg, Helv. Phys. Acta. 5, 369 (1932).

[19]If hyperfine coupling of nuclear spin $\vec{I}$ and electron angular momentum $\vec{J}$ is included, i.e., $j=F$, where $\vec{F}=\vec{I}+\vec{J}$, then the g factor is given by

$$g(F)=\tfrac{1}{2}g(J)\{1+[J(J+1)-I(I+1)]/F(F+1)]\} \quad (1b)$$

(neglecting nuclear magnetic moments); for Russell-Saunders coupling $g(J)$ is given by

$$g(J)=g_0\{\tfrac{3}{4}+\tfrac{1}{4}[S(S+1)-L(L+1)]/J(J+1)\} \quad (1c)$$

with $g_0\approx 2.00$. For hydrogen atoms in the $2\,^2S_{1/2}$ state with $F=1$, $g(F)\equiv g\equiv 1.00$.

[20]A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton U. P., Princeton, 1957), Chap. 4.

[21]R. T. Robiscoe, Am. J. Phys. 39, 146 (1971).

[22]For a discussion of the two-state curve crossing problem, see J. B. Delos and W. R. Thorson, Phys. Rev. A 6, 728 (1972).

[23]Cf. W. R. Thorson, J. B. Delos, and S. A. Boorstein, Phys. Rev. A 4, 1052 (1971); E. E. Nikitin, in *Fast Reactions and Primary Processes in Chemical Kinetics*, edited by S. Claesson (Almquist & Wiksells, Stockholm, 1967); see also E. E. Nikitin, in *Chemische Elementar Prozesse*, edited by H. Hartman (article in English) (Springer-Verlag, Berlin, 1968).

[24]J. Heinrichs, Phys. Rev. 176, 141 (1968).

[25]R. T. Robiscoe, *Cargese Lectures in Physics*, edited by M. Levy (Gordon and Breach, New York, 1968), pp. 3–53.