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## Use of Rotating Coordinates in Magnetic Resonance Problems

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The use of a rotating coordinate system to solve magnetic resonance problems is described. On a coordinate system rotating with the applied rotating magnetic field the effective field is reduced by the Larmor field appropriate to the rotational frequency. However, on such a coordinate system problems can more readily be solved since there is no time variation of the field. The solution in a stationary frame of reference is then obtained by a transformation from the rotating to the stationary frame. This procedure is equally valid in classical and in quantum-mechanical problems. The method is applied both to the molecular beam magnetic resonance method and to resonance absorption and nuclear induction experiments.

### I. INTRODUCTION

THE theory of the various molecular beam magnetic resonance methods and of the resonance absorption and nuclear induction experiments is usually chiefly concerned with the calculation of the effect of weak oscillating or rotating magnetic fields on nuclear magnetic moments in the presence of a strong constant magnetic field. Some of the simplest problems of this sort were first solved by Rabi,<sup>1</sup> Schwinger,<sup>2</sup> and Bloch<sup>3,4</sup> by a straightforward quantum-mechanical calculation of transition probabilities or by related methods. Although these methods are consistent with and closely related to the one described here, they are not as well suited to the simplified analysis of many more complicated problems. The extensive and explicit use of the rotating coordinate system was first developed by successive contributions of Bloch<sup>4</sup> and the present authors<sup>2</sup> over eight years ago. Since the method was originally considered chiefly as a new technique of calculation rather than as an intrinsically new result, no attempt was made to describe the method in the literature. However, in subsequent time it has become apparent that the value of the method in nuclear res-

onance calculations is so great that a detailed description of it is needed. This need is sufficiently great that several authors<sup>5-7</sup> have had to include a partial description of the rotating coordinate method in order to describe their experiments effectively.

The rotating coordinate system method is equally applicable to classical and quantum-mechanical systems. Because of the great simplicity and extensive applicability of the classical description it will be given first in detail while the quantum-mechanical case will be considered in the last section of this paper.

### II. CLASSICAL FORMULATION OF ROTATING COORDINATES PROCEDURE

Consider a system consisting of one or more nuclei or atoms all of which have the same constant gyromagnetic ratio  $\gamma$ . Then, if  $\mathbf{I}$  is the nuclear angular momentum in units of  $\hbar$ , the nuclear magnetic moment is  $\gamma\hbar\mathbf{I}$  and the equation of motion of the system in a stationary coordinate system is

$$\hbar \frac{d\mathbf{I}}{dt} = \mathbf{v} \times \mathbf{H} = \gamma\hbar\mathbf{I} \times \mathbf{H}. \quad (1)$$

But if  $\partial/\partial t$  represents differentiation with respect to a coordinate system that is rotating with angular

<sup>1</sup> I. I. Rabi, *Phys. Rev.* **51**, 652 (1937).

<sup>2</sup> J. Schwinger, *Phys. Rev.* **51**, 648 (1937); Rabi, Ramsey, and Schwinger, private communications and lecture notes (1945 on).

<sup>3</sup> F. Bloch and A. J. Siegert, *Phys. Rev.* **57**, 522 (1940); L. Alvarez and F. Bloch, *Phys. Rev.* **57**, 111 (1940).

<sup>4</sup> F. Bloch, *Phys. Rev.* **70**, 460 (1946); R. K. Wangsness and F. Bloch, *Phys. Rev.* **89**, 728 (1953).

<sup>5</sup> N. Bloembergen, *Nuclear Magnetic Relaxation* (Schotanus and Jens, Utrecht, Holland).

<sup>6</sup> H. C. Torrey, *Phys. Rev.* **76**, 1060 (1949).

<sup>7</sup> E. L. Hahn, *Phys. Rev.* **80**, 580 (1950).

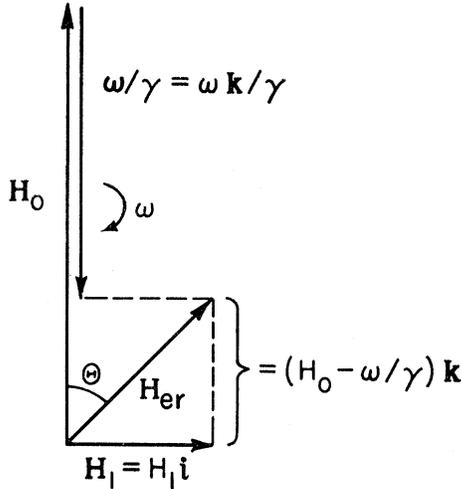


FIG. 1. Effective magnetic field in the rotating coordinate system.

velocity  $\omega$ ,

$$\frac{d\mathbf{I}}{dt} = \frac{\partial\mathbf{I}}{\partial t} + \omega \times \mathbf{I}, \quad (2)$$

where  $\mathbf{I}$  on both sides of the equation is the angular momentum as measured by the stationary observer, but the  $\partial\mathbf{I}/\partial t$  represents how a rotating observer would find the stationary observer's  $\mathbf{I}$  to vary as a function of time. By rearrangement Eq. (2) becomes

$$\hbar \frac{\partial\mathbf{I}}{\partial t} = \gamma \hbar \mathbf{I} \times (\mathbf{H} + \omega/\gamma) = \gamma \hbar \mathbf{I} \times \mathbf{H}_{er}, \quad (3)$$

where  $\mathbf{H}_{er}$  is the effective field in the rotating coordinate system and is defined by

$$\mathbf{H}_{er} = \mathbf{H} + \omega/\gamma. \quad (4)$$

In other words, the effect of the rotation of the coordinate system is merely to change the effective field by the added term  $\omega/\gamma$ .

This result can readily be applied to interpret the effect of the rotating magnetic fields used in the various nuclear resonance experiments. In most of these there is a constant field  $\mathbf{H}_0$  about which another (usually much weaker) field  $\mathbf{H}_1$  perpendicular to  $\mathbf{H}_0$  rotates with angular velocity  $-\omega$ . However, from the point of view of a coordinate system rotating with  $\mathbf{H}_1$ , none of the magnetic fields are changing as a function of time. Therefore, the axes of the rotating coordinate system can be selected so that

$$\mathbf{H}_0 = H_0 \mathbf{k}, \quad \mathbf{H}_1 = H_1 \mathbf{i}, \quad \omega = -\omega \mathbf{k}. \quad (5)$$

Then on the rotating coordinate system,

$$\mathbf{H}_{er} = (H_0 - \omega/\gamma) \mathbf{k} + H_1 \mathbf{i} \quad (6)$$

as is shown schematically in Fig. 1. Since this field is constant in time the solution to the motion of the

system is much simpler in the rotating coordinate system than in the stationary system. From Eq. (6) it follows that the magnitude of the effective magnetic field is

$$|\mathbf{H}_{er}| = [(H_0 - \omega/\gamma)^2 + H_1^2]^{\frac{1}{2}} = a/\gamma, \quad (7)$$

where

$$a = [(\omega_0 - \omega)^2 + (\gamma H_1)^2]^{\frac{1}{2}} = [(\omega_0 - \omega)^2 + (\omega_0 H_1/H_0)^2]^{\frac{1}{2}} \quad (8)$$

with  $\omega_0$  by definition being  $\gamma H_0$ . Likewise the angle  $\Theta$  of  $\mathbf{H}_{er}$  relative to  $\mathbf{H}_0$  is given by

$$\cos \Theta = (\omega_0 - \omega)/a, \quad \sin \Theta = (\omega_0 H_1/H_0)/a. \quad (9)$$

From this it is apparent that when  $\omega = \omega_0$ ,  $\Theta = 90^\circ$  and a magnetic moment initially parallel to  $\mathbf{H}_0$  can precess about  $\mathbf{H}_{er}$  until it becomes antiparallel to  $\mathbf{H}_0$ . In other words, such a moment can have its orientation relative to  $\mathbf{H}_0$  changed most completely when  $\omega = \omega_0$  so  $\omega_0$  can be considered as the resonance frequency of the system.

If one next goes to a second rotating coordinate system which rotates about  $\mathbf{H}_{er}$  with a suitable angular velocity, the effective field  $\mathbf{H}_{err}$  in the doubly rotating system can be reduced to zero. In this doubly rotating coordinate system the problems become trivial since there is then no magnetic field and consequently no change in the orientation of  $\mathbf{I}$ . If  $\mathbf{a}$  is the angular velocity about  $\mathbf{H}_{er}$  which reduces  $\mathbf{H}_{err}$  to zero,  $\mathbf{a}$  must be determined by

$$0 = \mathbf{H}_{err} = \mathbf{H}_{er} + \frac{\mathbf{a}}{\gamma}. \quad (10)$$

Hence, from (7) if  $\alpha$  is a unit vector parallel to  $\mathbf{H}_{er}$ ,

$$\mathbf{a} = -\gamma \mathbf{H}_{er} = -a\alpha, \quad (11)$$

where  $a$  is the quantity previously defined in Eq. (8).

As is shown in a later section of the present report, the above considerations apply to a quantum-mechanical as well as to a classical system. Consequently, either classically or quantum mechanically on the doubly rotating coordinate system with the two rotational angular velocities  $\omega$  and  $\mathbf{a}$ , there is no effective resultant field and the state of the system remains constant in time.

### III. CLASSICAL INTERPRETATION OF NUCLEAR RESONANCE EXPERIMENTS

The above can be directly applied to various problems arising in nuclear resonance experiments. Although oscillating instead of rotating fields are usually used in these experiments the problems can usually be treated as ones involving a rotating field since an oscillating field is equivalent to two opposite rotating fields and only the component rotating in direction to be able to give a resonance in Eq. (8) has an important effect in most problems.<sup>8</sup> As a first application the method can be used to demonstrate the criterion for the rate

<sup>8</sup> F. Bloch and A. J. Siegert, Phys. Rev. **57**, 522 (1940).

of change of a field to be "adiabatic," i.e., to be such that a nuclear moment preserves its magnetic quantum number (classically its angle) relative to the field as the field is moved. Let the field be rotated with angular velocity  $-\omega_1$ . Then for this problem, in the notation of the previous section,  $H_0$  is zero and  $H_1$  is the full field  $H$ . On the rotating coordinate system then

$$\mathbf{H}_{er} = (-\omega_1/\gamma)\mathbf{k} + H\mathbf{i}. \quad (12)$$

The nuclei will then preserve their orientation relative to  $H$  provided  $\mathbf{H}_{er}$  is approximately equal to  $H\mathbf{i}$  or that

$$|\omega_1| \ll H\gamma, \quad (13)$$

which can be written alternatively as

$$|\dot{\mathbf{H}} \times \mathbf{H}|/H^2 \ll \gamma H. \quad (14)$$

The use of the rotating coordinate system also provides a simple pictorial interpretation of the transition process that occurs in the molecular beam magnetic resonance method originally introduced by Rabi.<sup>9</sup> A singly rotating coordinate system rotating with the oscillator frequency  $-\omega$  can be used throughout. Prior to the molecule reaching the rotating field region  $H_1$  equals zero and  $\mathbf{H}_{er}$  has the value  $(H_0 - \omega/\gamma)\mathbf{k}$ . As the molecule enters the transition region where the rotating field is being established  $\mathbf{H}_{er}$  changes. Conditions are usually such that near resonance the condition of Eq. (14) applied to  $\mathbf{H}_{er}$  is violated. Consequently the transition under such circumstances is not adiabatic and the nuclear moments do not follow  $\mathbf{H}_{er}$  as  $H_1$  is established. After  $H_1$  achieves its full value  $\mathbf{H}_{er}$  is  $(H_0 - \omega/\gamma)\mathbf{k} + H\mathbf{i}$  and the nuclei precess about this effective field. When the molecules leave the rotating field region  $H_{er}$  again changes too rapidly for the nuclei to follow and they are left with the orientation relative to the  $z$  axis to which they have precessed in the region of the rotating field. At exact resonance this precession is about a field  $H_1$  which is perpendicular to the original direction of the field and consequently the change of orientation can be large.

The qualitative analysis of the preceding paragraph can be also expressed quantitatively. Assume the  $\mathbf{I}$  is initially parallel to  $\mathbf{H}_0$ . Then in the rotating coordinate system  $\mathbf{I}$  will precess about  $H_{er}$  with the precession angle  $\Theta$  and at an angular velocity  $a$ . If  $\alpha$  is the angle between  $\mathbf{H}_0$  and  $\mathbf{I}$ , the simple geometry of the above precession is such that after a time interval  $t_2 - t_1$ ,

$$\begin{aligned} \cos\alpha &= \cos^2\Theta + \sin^2\Theta \cos a(t_2 - t_1) \\ &= 1 - 2 \sin^2\Theta \sin^2 \frac{a(t_2 - t_1)}{1}. \end{aligned} \quad (15)$$

On the other hand, as shown in the next section, the quantum-mechanical angular momentum operators satisfy the same Eq. (1). Since this equation is linear,

the expectation values satisfy the same equation. Therefore, a correspondence between the classical and quantum-mechanical solutions can be established by requiring them to agree on their predicted average  $z$  component and on the total probability. If  $P_{\pm\frac{1}{2}}$  is the probability of a system of spin  $\frac{1}{2}$  being in the state of magnetic quantum number  $m$  equal to  $\pm\frac{1}{2}$  these requirements are

$$\begin{aligned} P_{\frac{1}{2}} - P_{-\frac{1}{2}} &= \cos\alpha, \\ P_{\frac{1}{2}} + P_{-\frac{1}{2}} &= 1. \end{aligned} \quad (16)$$

Therefore,

$$\begin{aligned} P_{-\frac{1}{2}} &= \frac{1 - \cos\alpha}{2} = \sin^2\Theta \sin^2 \frac{a(t_2 - t_1)}{2} \\ &= \frac{(\omega_0 H_1 / H_0)^2}{(\omega_0 - \omega)^2 + (\omega_0 H_1 / H_0)^2} \sin^2 \frac{a(t_2 - t_1)}{2}. \end{aligned} \quad (17)$$

As proved in Sec. V, this is exactly the same result that is obtained from a pure quantum-mechanical calculation.

Likewise the rotating coordinate system analysis procedure is applicable to the molecular beam resonance method with separated oscillating fields introduced by Ramsey.<sup>10</sup> In this case, the description through the first oscillating field is just the same as in the preceding paragraph. After leaving the oscillating field in this method the nucleus enters an intermediate region where there is only  $H_0$  so the magnitude of  $H_1$  is zero. Relative to the singly rotating coordinate system, the nucleus in this region precesses about  $(H_0 - \omega/\gamma)\mathbf{k}$  until it reaches the second oscillating field. As a result of this precession the nucleus will in general have a different orientation relative to  $\mathbf{H}_{er}$  in the second rotating field region than it did in the first. On the other hand, if the average value of  $H_0 - \omega/\gamma$  in the intermediate region is zero the orientation of the nucleus relative to  $\mathbf{H}_{er}$  is the same in the second oscillating field region as in the first. This will be true regardless of the velocity of the molecule. However, if the average of  $H_0 - \omega/\gamma$  has any value other than zero, the orientation of the nucleus relative to  $\mathbf{H}_{er}$  in the second field is different from that in the first and the magnitude of the difference will depend upon the velocity of the molecule. When the combined effects of the two rotating field regions are averaged over the velocity of the molecules, it is therefore found that the transition probability is a maximum for  $\omega$  equal to the average value of  $\gamma H_0$  in the intermediate region.

The rotating coordinate system procedure is also of value in interpreting the various nuclear resonance absorption and induction experiments based on the original experiments of Purcell<sup>11</sup> and Bloch.<sup>12</sup> Experiments of these types have by now been carried out in a number of different ways. One of the marked differences has been

<sup>9</sup> Kusch, Millman, and Rabi, Phys. Rev. **55**, 666 (1939); Kellogg, Rabi, Ramsey, and Zacharias, Phys. Rev. **55**, 318 (1939).

<sup>10</sup> N. F. Ramsey, Phys. Rev. **78**, 695 (1950).

<sup>11</sup> Purcell, Torrey, and Pound, Phys. Rev. **69**, 37 (1946).

<sup>12</sup> Bloch, Hansen, and Packard, Phys. Rev. **69**, 127 (1946).

in the extent to which the adiabatic condition of Eq. (14) is satisfied for  $\mathbf{H}_{er}$ . In some experiments a large value of the rotating field  $H_1$  has been used and the field  $H_0$  has been varied about the resonance value at a sufficiently slow rate that Eq. (14) was satisfied. In this case the original magnetism,

$$\mathbf{M}_0 = \chi \mathbf{H}_0, \quad (18)$$

simply retains its orientation relative to  $\mathbf{H}_{er}$  in the singly rotating coordinate system and the dominant effect of the small changes in  $H_0$  is to change the orientation of  $\mathbf{H}_{er}$  and hence  $\mathbf{M}_0$ . Therefore the  $x$  component of this in the rotating system varies with  $H_0$  as

$$M_x = M_0 \cos\theta = \frac{M_0 \omega_0 H_1 / H_0}{[(\omega_0 - \omega)^2 + (\omega_0 H_1 / H_0)^2]^{\frac{1}{2}}}. \quad (19)$$

$M_x$  then goes through a resonance maximum at  $\omega = \omega_0$ . However, as the coordinate system and hence  $M_x$  are rotating with frequency  $\omega$  relative to the laboratory system, the resulting rotating magnetic moment will induce a signal in a suitably placed coil.

Actually the preceding case rarely applies since the local magnetic fields from neighboring molecules cause the nuclei to return to the thermal equilibrium relative to  $\mathbf{H}_0$  and to lose their phase coherency about  $\mathbf{H}_0$ . The characteristic time constants for these processes are conventionally designated  $T_1$  and  $T_2$ .<sup>4</sup> From the point of view of the singly rotating frame of reference,  $T_1$  is the characteristic time for resuming thermal equilibrium about the  $z$  axis, and  $T_2$  is the characteristic time constant for losing coherency about that axis, i.e., for the  $x$  and  $y$  components of the magnetization to average to zero. Although the preceding results are not directly applicable when these relaxation effects are important, they are nevertheless of value in many approximate calculations even then. For example, if  $T_2 \ll T_1$ ,  $T_2 \ll 1/\gamma H_1$ , and  $(T_1 T_2)^{\frac{1}{2}} \ll 1/\gamma H_1$ , the rate of absorption of energy at resonance in a nuclear resonance absorption experiment with nuclei of spin  $\frac{1}{2}$  can roughly be approximated by the assumption that the transition goes as in Eq. (17) but is stopped by the loss of all coherency after a time  $T_2$ . At exact resonance and for a time  $T_2$ , Eqs. (17) and (8) give for the transition probability

$$P_{-\frac{1}{2}} = \sin^2(\gamma H_1 T_2 / 2) \approx \frac{1}{4} \gamma^2 H_1^2 T_2^2. \quad (20)$$

The average rate  $W$  of transition per nucleus is then approximately given by

$$W = P_{-\frac{1}{2}} / T_2 = \frac{1}{4} \gamma^2 H_1^2 T_2. \quad (21)$$

Therefore, the average rate  $R$  of net absorption of energy is approximated by

$$R = W n_s \hbar \omega = (1/8) \gamma^2 H_1^2 T_2 N \hbar^2 \omega^2 / kT, \quad (22)$$

where  $n_s = \frac{1}{2} N \hbar \omega / kT$  is the number of surplus nuclei in the lowest energy state,  $N$  is the total number of nuclei present,  $\hbar \omega$  is the energy separation of the two orientation states, and  $T$  is the absolute temperature.

Other limiting cases with the various relaxation processes have been discussed in the literature.<sup>4-6,13</sup> As has been discussed by Hahn,<sup>7</sup> the rotating coordinate system procedure is particularly well suited to the description of the spin echo phenomena.

#### IV. QUANTUM-MECHANICAL FORMULATION OF ROTATING COORDINATES PROCEDURE

Relation (4) derived above can be proved equally well quantum mechanically as classically. As indicated in the previous section, one procedure is simply to say that Eq. (1) applies to the quantum-mechanical angular momentum operators. Equation (1) can in fact easily be proved to follow from the standard operator relation

$$(\hbar/i) d\mathbf{I}/dt = [\mathcal{H}, \mathbf{I}] = \mathcal{H}\mathbf{I} - \mathbf{I}\mathcal{H}, \quad (23)$$

where the Hamiltonian  $\mathcal{H}$  is taken as

$$\mathcal{H} = -\gamma \hbar \mathbf{I} \cdot \mathbf{H}. \quad (24)$$

Alternatively, from a wave mechanical point of view, the Schrödinger equation for the problem relative to a stationary coordinate system is

$$i\hbar \dot{\Psi} = \mathcal{H}\Psi = -\gamma \hbar \mathbf{I} \cdot \mathbf{H}\Psi. \quad (25)$$

However, in quantum mechanics the finite rotation operator<sup>14</sup> for the coordinates to be rotated an angle  $\zeta$  about an axis along which the system's angular momentum,  $\mathbf{I}$ , has the component  $I_\zeta$  is the unitary operator  $\exp(i\zeta I_\zeta)$ . Let  $\Psi$  and  $\mathbf{H}$  be the wave function and field relative to a stationary coordinate system whereas  $\Psi_r$  and  $\mathbf{H}_r$  are the same quantities relative to coordinates rotating with angular velocity  $\omega$ . These quantities are related by the unitary transformation so that<sup>14</sup>

$$\Psi = \exp(-i\omega \cdot \mathbf{I}t) \Psi_r, \quad (26)$$

$$\mathbf{I} \cdot \mathbf{H}_r = \exp(i\omega \cdot \mathbf{I}t) \mathbf{I} \cdot \mathbf{H} \exp(-i\omega \cdot \mathbf{I}t). \quad (27)$$

If Eq. (26) is substituted in Eq. (25) and if Eq. (27) is used to simplify the resulting expression one immediately obtains

$$i\hbar \dot{\Psi}_r = -\gamma \hbar \mathbf{I} \cdot (\mathbf{H}_r + \omega/\gamma) \Psi_r = -\gamma \hbar \mathbf{I} \cdot \mathbf{H}_{er} \Psi_r, \quad (28)$$

where  $\mathbf{H}_{er}$  is given by Eq. (4), with the understanding that  $\mathbf{H}$  is to be expressed relative to the rotating coordinate system. This result justifies the application of the previous discussions to quantum-mechanical as well as classical systems.

#### V. QUANTUM-MECHANICAL CALCULATION OF TRANSITION PROBABILITY

The probabilities for transition of the system from a state of one magnetic quantum number to another can readily be calculated with the above procedure. Consider the case discussed earlier in which the mag-

<sup>13</sup> Bloembergen, Purcell, and Pound, *Phys. Rev.* **73**, 679 (1948).

<sup>14</sup> E. C. Kemble, *Fundamental Principles of Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York), pp. 247, 307, and 532.

netic fields are given by Eq. (5) and assume that up to time  $t_1$  the magnitude of  $H_1$  is zero after which it is  $H_1$  until time  $t_2$ . As use will be made of the stationary, singly, and doubly rotating coordinate systems previously discussed, wave functions relative to these three systems will be designated as  $\Psi(t)$ ,  $\Psi_r(t)$ , and  $\Psi_{rr}(t)$ , respectively. Since  $\mathbf{H}_{err}$  for the doubly rotating coordinates is zero,

$$\Psi_{rr}(t_2) = \Psi_{rr}(t_1) = \Psi_r(t_1). \quad (29)$$

However, as the doubly rotating system between times  $t_1$  and  $t_2$  has rotated an angle  $-a(t_2-t_1)$  relative to the singly rotating system one must use the previous finite rotation operator to relate  $\Psi_{rr}(t_2)$  and  $\Psi_r(t_2)$  with the result that

$$\Psi_r(t_2) = \exp(ia[t_2-t_1]\boldsymbol{\alpha} \cdot \mathbf{I})\Psi_{rr}(t_2). \quad (30)$$

Hence from (29),

$$\Psi_r(t_2) = \exp(ia[t_2-t_1]\boldsymbol{\alpha} \cdot \mathbf{I})\Psi_r(t_1). \quad (31)$$

In a similar fashion, this can be reduced to a non-rotating coordinate system with the result

$$\Psi(t_2) = \exp(i\omega t_2 \mathbf{k} \cdot \mathbf{I}) \exp(ia[t_2-t_1]\boldsymbol{\alpha} \cdot \mathbf{I}) \times \exp(-i\omega t_1 \mathbf{k} \cdot \mathbf{I}) \Psi(t_1). \quad (32)$$

It should be noted, however, that because of the components of  $\mathbf{I}$  not commuting, one cannot perform all the operations appropriate to exponentials of ordinary numbers; instead, the exponentials may be taken as defined by their series expansion.

From the above, the transition probability from a state  $m$  to a state  $m'$  can be calculated by taking  $\Psi(t_1) = \Psi_m$  in which case

$$P_{m' \leftarrow m} = |\langle \Psi_{m'}, \Psi(t) \rangle|^2 = \langle m' | \exp(i\omega t_2 \mathbf{k} \cdot \mathbf{I}) \times \exp(ia[t_2-t_1]\boldsymbol{\alpha} \cdot \mathbf{I}) \exp(-i\omega t_1 \mathbf{k} \cdot \mathbf{I}) | m \rangle|^2 = \langle m' | \exp(ia[t_2-t_1]\boldsymbol{\alpha} \cdot \mathbf{I}) | m \rangle|^2. \quad (33)$$

The last simplifying step is a consequence of  $\Psi_m$  and  $\Psi_{m'}$  being eigenfunctions of  $\mathbf{k} \cdot \mathbf{I}$ .

In general, the numerical evaluation of (33) is somewhat cumbersome because of the noncommutativity of the terms in the exponent. However, a series expansion of the exponential may be used. In the special case of spin  $\frac{1}{2}$ , it becomes much simplified for then  $\mathbf{I} = \frac{1}{2}\boldsymbol{\sigma}$ , where  $\boldsymbol{\sigma}$  is the Pauli spin matrix. Since for the Pauli spin matrix  $(\boldsymbol{\alpha} \cdot \boldsymbol{\sigma})^2$  equals one, the series expansion of the above exponential together with the series expansion for sine and cosine merely give

$$\begin{aligned} & \exp\left(\frac{a}{2}[t_2-t_1]\boldsymbol{\alpha} \cdot \boldsymbol{\sigma}\right) \\ &= \cos\frac{a(t_2-t_1)}{2} + i\boldsymbol{\alpha} \cdot \boldsymbol{\sigma} \sin\frac{a(t_2-t_1)}{2} \\ &= \cos\frac{a(t_2-t_1)}{2} + i(\sigma_x \sin\Theta + \sigma_z \cos\Theta) \sin\frac{a(t_2-t_1)}{2}. \end{aligned} \quad (34)$$

Therefore

$$P_{-\frac{1}{2} \leftarrow \frac{1}{2}} = \sin^2\Theta \sin^2\frac{a(t_2-t_1)}{2}, \quad (35)$$

which is the desired transition probability that is applicable to the conventional molecular beam resonance method.<sup>9</sup> It should be noted that the above agrees with the classically derived expression in Eq. (17).

This procedure may also be used to calculate the transition probability applicable to the molecular beam resonance method with separated oscillating fields.<sup>10</sup> In this case, Eq. (32) can be applied separately to the first oscillating field region, to the intermediate region, and to the second oscillating field region. The resulting three equations can then be combined to express the final state of the system in terms of the initial state and from this the transition probabilities can be calculated as in Eq. (25).

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