Folded Stern-Gerlach Experiment as a Means for Detecting Nuclear Magnetic Resonance in Individual Nuclei

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In a Stern-Gerlach experiment, the spatial trajectory of a particle traversing a magnetic-field gradient serves as a detector of its spin state. This paper considers the implications of adding a harmonic restoring potential to the Stern-Gerlach experiment. The behavior of such systems is shown to resemble that of a "folded" Stern-Gerlach experiment, in that the linear spatial trajectory of the Stern-Gerlach experiment becomes folded into the cyclic phase-space trajectory of the oscillator. Under appropriate circumstances, the coupling between the spin and the oscillator is shown to be sufficiently strong that it might find practical application as the basis of a magnetic resonance imaging device.

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Consider a system made up of three elements: (1) a harmonic oscillator, (2) a spin-$\frac{1}{2}$ particle (nominally a proton) mounted on the oscillator, and (3) an externally applied magnetic field. The magnetic moment of the particle will create a coupling between the field and the oscillator. How strong is the coupling? What is the dynamic behavior of the coupled system? This paper shows that, under the proper conditions, the coupling is sufficiently strong that magnetic resonance imaging of a single nucleus may be feasible, by monitoring the excitation of an oscillator coupled to the nucleus.

Let ω be the resonant frequency of the mechanical oscillator, and m the oscillator mass. We adopt units $\hbar=\omega=m=1$. The external magnetic field $\mathbf{B}(\mathbf{x},t)$ is assumed to have a uniform component, a time-dependent component, and a gradient component;

$$\mathbf{B}(\mathbf{x},t)=\mathbf{B}_c+\mathbf{B}_t\cos(t)+\mathsf{G}\cdot\mathbf{x}\,, \tag{1}$$

with $\mathbf{B}_c$, $\mathbf{B}_t$, and G constant. The Hamiltonian of the coupled spin-oscillator system is

$$H=\tfrac{1}{2}(p^2+q^2)-\gamma\mathbf{B}_c\cdot\mathbf{I}-\gamma\mathbf{B}_t\cdot\mathbf{I}\cos(t)-\gamma q\hat{\mathbf{n}}\cdot\mathsf{G}\cdot\mathbf{I}\,. \tag{2}$$

Here p and q are oscillator momentum and position operators, satisfying $[q,p]=i$. The unit vector $\hat{\mathbf{n}}$ is the axis of oscillator motion. The spin operators $\mathbf{I}=\{I_x,I_y,I_z\}$ satisfy $[I_x,I_y]=iI_z$. With this convention I_z has eigenvalues $\pm\frac{1}{2}$. The gyromagnetic ratio γ gives the spin precession frequency at a given field strength.

This Hamiltonian is identical to that of Stern-Gerlach-Rabi experiments, except for the added harmonic potential $\frac{1}{2}q^2$. From inspection of the Hamiltonian, it is evident that the spin undergoes Rabi-type precession in response to a time-dependent field which has two components, namely, the externally applied field $\mathbf{B}_t\cos(t)$, and the oscillator-generated field $q\hat{\mathbf{n}}\cdot\mathsf{G}$. As usual in analyzing Rabi precession [1], we move to an interaction picture with the zero-order Hamiltonian $H_0=\frac{1}{2}(p^2+q^2)-\hat{\mathbf{z}}\cdot\mathbf{I}$. Here $\hat{\mathbf{z}}$ is a unit vector in the direction of the local field $\mathbf{B}_c$. The interaction-picture Hamiltonian is of the form

$$H_{\rm int}=-\gamma\mathbf{B}_{\rm eff}\cdot\mathbf{I}_{\rm rot}\,, \tag{3}$$

with $\mathbf{B}_{\rm eff}$ an effective field as seen in a reference frame rotating about $\hat{\mathbf{z}}$ with unit angular velocity. The rotating-frame spin $\mathbf{I}_{\rm rot}$ is defined by

$$\mathbf{I}\equiv[\hat{\mathbf{z}}\otimes\hat{\mathbf{z}}+\cos(t)(\mathsf{I}-\hat{\mathbf{z}}\otimes\hat{\mathbf{z}})+\sin(t)\hat{\mathbf{z}}^*]\mathbf{I}_{\rm rot}\,. \tag{4}$$

Here $\hat{\mathbf{z}}^*$ is the antisymmetric matrix $(\hat{z}^*)_{jk}\equiv\hat{z}_i\varepsilon_{ijk}$. Henceforth we work exclusively in the rotating frame, and drop the subscript from $\mathbf{I}_{\rm rot}$.

It is convenient to parametrize $\mathbf{B}_{\rm eff}$ in Eq. (3) in the following standard way:

$$\mathbf{B}_{\rm eff}=\frac{1}{\gamma}\left(\frac{X-L_x}{2L}\hat{\mathbf{x}}+\frac{Y-L_y}{2L}\hat{\mathbf{y}}+\delta\hat{\mathbf{z}}\right). \tag{5}$$

Here X and Y are oscillator amplitude operators

$$\begin{bmatrix}X\\Y\end{bmatrix}=\begin{bmatrix}\cos(t)&-\sin(t)\\\sin(t)&\cos(t)\end{bmatrix}\begin{bmatrix}q\\p\end{bmatrix}, \tag{6}$$

satisfying $[X,Y]=i$. Classically, X and Y are the in-phase and quadrature components of the oscillator amplitude. The length scale L and the unit vectors $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are defined in terms of the field gradient:

$$\hat{\mathbf{x}}/L=\gamma(\mathsf{I}-\hat{\mathbf{z}}\otimes\hat{\mathbf{z}})\cdot\mathsf{G}\cdot\hat{\mathbf{n}}\,, \tag{7}$$

$$\hat{\mathbf{y}}=\hat{\mathbf{z}}\times\hat{\mathbf{x}}\,. \tag{8}$$

The lengths L_x and L_y parametrize the drive field $\mathbf{B}_t$:

$$L_x/L=-\gamma\mathbf{B}_t\cdot\hat{\mathbf{x}}\,, \tag{9}$$

$$L_y/L=-\gamma\mathbf{B}_t\cdot\hat{\mathbf{y}}\,. \tag{10}$$

The physical interpretation of L_x and L_y is as follows. We ask, for what excitation of the oscillator does the oscillator-generated field cancel the transverse component of the applied field $\mathbf{B}_t$? By Eq. (5), this occurs at $X=L_x$, $Y=L_y$.

The dimensionless tuning parameter δ in Eq. (5) is

$$\delta=\gamma|\mathbf{B}_c|-1\,, \tag{11}$$

so that δ vanishes when the spin precession frequency $\gamma|\mathbf{B}_c|$ matches the oscillator frequency $\omega=1$.

We begin by treating the oscillator and spin motion purely classically, i.e., regarding X, Y, and $\mathbf{I}$ in Eqs. (3)–(5) as classical fields.

The equation of motion for $\langle\mathbf{I}\rangle$ is

$$\frac{d\langle\mathbf{I}\rangle}{dt}=-\gamma\mathbf{B}_{\rm eff}\times\langle\mathbf{I}\rangle\,. \tag{12}$$

This is the expected result; $\mathbf{B}_{\rm eff}$ generates Rabi precession of the spins. The spin vector $\mathbf{I}$ can be separated into components $\mathbf{I}_{\rm par}$ and $\mathbf{I}_{\rm perp}$, parallel to and perpendicular to $\mathbf{B}_{\rm eff}$. The adiabatic theorem [2] implies that if the system starts in a state with $\langle\mathbf{I}_{\rm par}\rangle=m_s\hat{\mathbf{B}}_{\rm eff}$, then $\langle\mathbf{I}_{\rm par}\rangle=m_s\hat{\mathbf{B}}_{\rm eff}$ at all later times as well, so that m_s is an (approximate) constant of the motion.

In the classical equation of motion for X and Y, terms proportional to $\langle\mathbf{I}_{\rm perp}\rangle$ oscillate with the Rabi frequency $\gamma|\mathbf{B}_{\rm eff}|$, and can be neglected in a secular approximation. Thus $\langle\mathbf{I}\rangle\approx\langle\mathbf{I}_{\rm par}\rangle\approx m_s\hat{\mathbf{B}}_{\rm eff}$ and we obtain

$$\frac{d}{dt}\begin{bmatrix}X\\Y\end{bmatrix}=\begin{bmatrix}0 & -\omega_{\rm orbit}(X,Y)\\ \omega_{\rm orbit}(X,Y) & 0\end{bmatrix}\begin{bmatrix}X-L_x\\Y-L_y\end{bmatrix}, \tag{13}$$

where the orbital frequency $\omega_{\rm orbit}$ is

$$\omega_{\rm orbit}(X,Y)=m_s\frac{1}{2L}[4\delta^2L^2+(X-L_x)^2+(Y-L_y)^2]^{-1/2}. \tag{14}$$

This equation can be solved by inspection, once we notice that $\omega_{\rm orbit}$ is a constant of the motion. The (undamped) oscillator trajectory in phase space is a circular orbit about the point $\{L_x,L_y\}$ with uniform angular velocity $\omega_{\rm orbit}$.

We can check these classical results by solving the quantum equations exactly. It is convenient to introduce three different basis sets. The initial state is conveniently specified in the familiar "harmonic" basis $\{|n,\uparrow\rangle_h,\ |n,\downarrow\rangle_h;\ n=0,1,2,\ldots\}$, which is generated by the usual raising and lowering operators. The time evolution is computed by transforming to a shifted basis (defined below) $\{|n,\uparrow\rangle_s,\ |n,\downarrow\rangle_s;\ n=0,1,2,\ldots\}$, and then further transforming to an eigenstate basis (also defined below)

$$\{|n,+\rangle_e,|n,-\rangle_e\,;\,n=0,1,2,\ldots\}\,.$$

The shifted basis is defined in terms of the shifted amplitude operators $X_s\equiv X-L_x$ and $Y_s\equiv Y-L_y$, and the shifted raising and lowering operators $a_s^{\pm}\equiv(X_s\mp iY_s)/\sqrt{2}$. Then $[a_s^-,a_s^+]=1$ (as usual). The Hamiltonian takes a simple form in the shifted basis

$$H_{\rm int}=-\left(\frac{1}{2L\sqrt{2}}(a_s^+I^++a_s^-I^-)+\delta I_z\right). \tag{15}$$

The eigenstates $|n,\pm\rangle_e$ of $H_{\rm int}$ occur in pairs with opposite-sign eigenvalues $\lambda_n^{\pm}$, labeled by an integer quantum number n, such that

$$H_{\rm int}|n,\pm\rangle_e=\lambda_n^{\pm}|n,\pm\rangle_e\,, \tag{16}$$

and

$$|n,\pm\rangle_e=\frac{|n,\uparrow\rangle_s-\alpha_{\pm}|n-1,\downarrow\rangle_s}{(1+\alpha_{\pm}^2)^{1/2}}\,. \tag{17}$$

The mixing amplitude $\alpha_{\pm}$ and eigenvalues $\lambda_n^{\pm}$ are

$$\alpha_{\pm}=L(2\lambda_n^{\pm}+\delta)/\sqrt{n/2}\,, \tag{18}$$

$$\lambda_n^{\pm}=\pm\tfrac{1}{2}(\delta^2+n/2L^2)^{1/2}. \tag{19}$$

The shifted basis does not contain $|-1,\downarrow\rangle_s$, so the $|0,+\rangle_e$ eigenstate is not defined. The basis set $\{|n,\pm\rangle_e\}$ is complete even without $|0,+\rangle_e$.

Now suppose a wave packet is constructed as a superposition of states $|n,\pm\rangle_e$, with mean quasienergy quantum number $\langle n_e\rangle$. A wave packet localized at $\{X,Y\}$ has energy quantum numbers centered about

$$\langle n_e\rangle\approx\tfrac{1}{2}[(X-L_x)^2+(Y-L_y)^2]\,. \tag{20}$$

Expanding the n dependence of $\lambda_n^{\pm}$ to first order yields

$$\left.\frac{d\lambda_n^{\pm}}{dn}\right|_{n=\langle n_e\rangle}=\pm\,\omega_{\rm orbit}\,. \tag{21}$$

Because n is an integer, quantum wave packets therefore have (to leading order) a periodic time dependence $\omega_{\rm orbit}$, as was also obtained classically.

To display the time evolution of a wave packet, it is necessary to take into account that X and Y are noncommuting operators. We introduce a basis set of coherent oscillator states $|x,y\rangle$ defined by

$$a_h^-|x,y\rangle\equiv(x+iy)|x,y\rangle/\sqrt{2}\,. \tag{22}$$

Here x and y are ordinary numbers which label the state, not operators. Defined in this way, the coherent states $|x,y\rangle$ are not orthogonal. However, they satisfy a pseudo-orthogonality relation [1]

$$|\phi\rangle=\frac{1}{\pi}\int_{-\infty}^{-\infty}dx\int_{-\infty}^{-\infty}dy|x,y\rangle\langle x,y|\phi\rangle\,, \tag{23}$$

where $|\phi\rangle$ is an arbitrary state. The wave function $\psi(x,y,t)$ defined by

$$\psi(x,y,t)\equiv\langle x,y|\psi(t)\rangle \tag{24}$$

therefore is a complete description of the oscillator state, because knowledge of $\psi(x,y,t)$ allows us to expand $|\psi(t)\rangle$ in any other basis.

Physically, $\psi(x,y,t)$ gives the amplitude for the oscillator to be found in a localized state $|x,y\rangle$ with $\langle X\rangle=x$ and $\langle Y\rangle=y$.

Figure 1 shows the evolution of a quantum wave packet initialized with the oscillator in its ground state (i.e.,

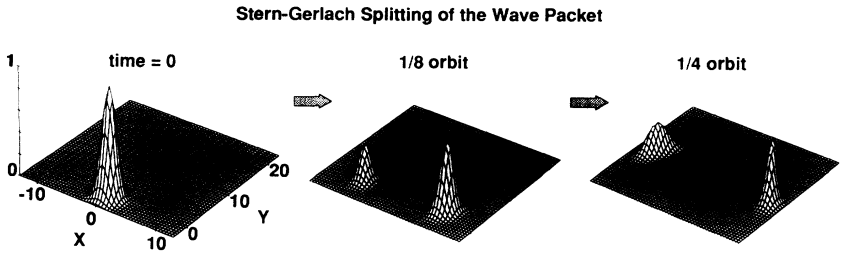

FIG. 1. Stern-Gerlach splitting of the wave packet.

maximally localized at $\langle X\rangle=0$ and $\langle Y\rangle=0$). The plotted quantity in Fig. 1 is $|\psi(x,y,t)|^2$, summed over spin quantum numbers. The drive field $\mathbf{B}_t$ is such that $L_x=0$ and $L_y=10$ (in units of length $\sqrt{\hbar/m\omega}$).

To demonstrate Stern-Gerlach separation of the wave packet, the initial spin state in Fig. 1 is deliberately chosen to be oblique to $\hat{\mathbf{B}}_{\rm eff}=-\hat{\mathbf{y}}$. Specifically, $\langle\mathbf{I}\rangle$ is chosen so that

$$\langle\mathbf{I}\rangle=-\tfrac{1}{2}(\tfrac{1}{3}\hat{\mathbf{x}}+\sqrt{8/9}\hat{\mathbf{y}})\,. \tag{25}$$

Thus the initial packet contains both negative-frequency eigenstates $|n,-\rangle_e$ and positive-frequency eigenstates $|n,+\rangle_e$, in $\frac{2}{3}$ to $\frac{1}{3}$ proportion.

Under time evolution, an inhomogeneous packet of this type does not remain localized. Instead, it divides into separate positive-frequency and negative-frequency wave packets, which follow separate trajectories in X-Y phase space.

This has the important practical consequence that the oscillator becomes mechanically excited regardless of the initial spin state. It is unnecessary to specially prepare the spin state (e.g., with a "$\pi/2$" pulse as in magnetic resonance imaging).

An analogous packet separation occurs in a Stern-Gerlach-Rabi apparatus. In the Stern-Gerlach experiment, an arbitrary initial wave packet can be decomposed into up-spin and down-spin packets, whose trajectories subsequently separate in q-p phase space. Similarly, in the present case an arbitrary initial packet can be decomposed into an $|n,+\rangle_e$ packet and a $|n,-\rangle_e$ packet. These two packets are down-spin and up-spin relative to the rotating-frame field $\mathbf{B}_{\rm eff}$. Packet separation subsequently occurs in X-Y phase space, as shown in Fig. 1.

It follows that, in the classical equations of motion [Eqs. (13) and (14)], m_s can have only the discrete values $\pm\frac{1}{2}$. Other values of m_s are classically possible, but are not supported by the underlying quantum mechanics, just as in the Stern-Gerlach experiment.

Oscillator damping can be modeled by adding an Ohmic damping term to the X-Y equations of motion:

$$\frac{d}{dt}\begin{bmatrix}X\\Y\end{bmatrix}=\begin{bmatrix}0&-\omega_{\rm orbit}\\ \omega_{\rm orbit}&0\end{bmatrix}\begin{bmatrix}X-L_x\\Y-L_y\end{bmatrix}+\begin{bmatrix}-1/2Q&0\\0&-1/2Q\end{bmatrix}\begin{bmatrix}X\\Y\end{bmatrix}. \tag{26}$$

Here Q is the quality factor of the oscillator.

The equilibrium solutions of the damped equations represent states with continuous power flow $\langle n\rangle/Q$ into the oscillator. It can be shown that a sufficient condition for a unique, stable equilibrium to exist is that $\mathbf{B}_t$ satisfy

$$\langle n\rangle/Q\le\tfrac{1}{4}\gamma|(\mathbf{1}-\hat{\mathbf{z}}\otimes\hat{\mathbf{z}})\cdot\mathbf{B}_t|\,. \tag{27}$$

Classically, this is equivalent to the requirement that $\mathbf{B}_t$ couple enough power into the oscillator to balance dissipative losses.

The occupation number $\langle n\rangle$ of the oscillator at equilibrium (in the harmonic basis) is found to be

$$\langle n\rangle=\tfrac{1}{4}\{4\delta^2L^2+L_{\rm max}^2+L_x^2+L_y^2-[(L_{\rm max}^2+L_x^2+L_y^2+4\delta^2L^2)^2-4L_{\rm max}^2(L_x^2+L_y^2)]^{1/2}\}\,. \tag{28}$$

Here $L_{\rm max}=Q/2L$ is the upper bound on the oscillator amplitude which can be driven by a single spin. In terms of $L_{\rm max}$, the external power requirement on $\mathbf{B}_t$ [Eq. (27)] is equivalent to

$$L_x^2+L_y^2\ge L_{\rm max}^2\,. \tag{29}$$

In physical terms, the field externally applied to the particle should be larger than the maximum possible oscillator-generated field.

Thermal fluctuations in the oscillator can induce transitions between positive-frequency and negative-frequency states. To calculate the rate $R_{\rm flip}$ at which a positive-frequency packet decays into a negative-frequency packet, the thermal environment is modeled by a Langevin force term in the Hamiltonian. The coupled spin-oscillator system is assumed to be in equilibrium with $\delta=0$, as per Eq. (28). Using the fluctuation-dissipation theorem to determine the spectral density of oscillator fluctuations as a function of temperature T, and applying first-order time-dependent perturbation theory, leads to

the following expression for R_{flip}:

$$R_{\text{flip}} = \frac{1}{Q} \frac{L_{\max}^2}{L_x^2 + L_y^2 - L_{\max}^2} \frac{kT}{8\langle n \rangle} . \quad (30)$$

Here k is Boltzmann's constant. So if the excitation of the oscillator is above thermal noise, and $\mathbf{B}_f$ is sufficiently strong that $L_x^2 + L_y^2 \gg L_{\max}^2$, then the probability of a thermally induced spin slip occurring during an oscillator damping time $1/Q$ is small.

Oscillator-based NMR detection differs substantially from inductive NMR detection, and it is necessary to "unlearn" some conventional wisdom regarding inductive NMR. In oscillator-based detection it is not necessary to polarize samples prior to taking a measurement, nor is it necessary to wait for a period $\sim T_1$ after each measurement for the polarization to equilibriate. $\pi/2$ pulses are not required to initiate spin precession, because any initial spin state couples energy into the oscillator.

The novel characteristics of oscillator-based NMR detection arise from the appearance of field gradients in a new and unfamiliar role, as a source of mechanical force, rather than as a means of modulating spin phases.

Another paper [3] describes some specific designs for oscillator-based detectors. The predicted single-nucleon signals lie well above quantum and thermodynamic detection limits. The intrinsic spatial resolution is of order $(L_x^2 + L_y^2)^{1/2}$, which for a suitable design can be less than an angstrom.

Bloom and co-workers have also described a resonant Stern-Gerlach experiment [4,5]. Their approach is similar to that described here, in that they observed Stern-Gerlach separation of the wave packet occurring in a rotating frame of reference associated with a time-dependent applied field. Their experiment differed in that the gradient was time dependent, rather than static as in the present case, and the trajectory of the particles was spatially extended, rather than folded as in the present case.

There is considerable scope for invention in oscillator-based NMR detection. Alternative and better instrument designs might well differ radically from those presented in [3]. Readers are encouraged to exercise their talents in this area.

There is an urgent medical need for the information that a molecular imaging device could provide. Amino acid sequences are known for thousands of biologically important molecules, but we do not know the structure of these molecules or how they work, except for a few exceptional cases. This makes the rational design of treatments (e.g., for the HIV disease) very slow and difficult.

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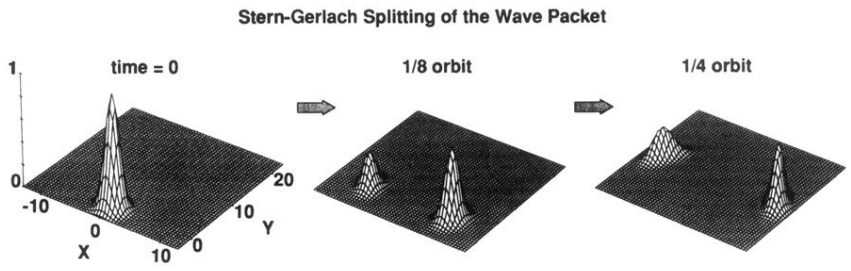

FIG. 1. Stern-Gerlach splitting of the wave packet.