Dynamic localization in spin systems

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(Received 9 September 1999)

A dilute collection of spins, when driven coherently by crossed magnetic fields of arbitrary and controllable time dependences along the three Cartesian directions, is shown to exhibit a striking phenomenon in which the azimuthal quantum number remains unchanged for certain resonant combinations of the field intensities and field frequencies. The formalism of dynamic localization available for the area of charge transport in crystals is shown to provide an approximate but highly efficient analytic method for the study of this phenomenon. The effect on scattering of probe particles is elucidated.

Quantum tunneling of magnetization has become an intensely studied field in various systems of physics and chemistry, [1–7] in part because quantum effects, normally difficult to observe in macroscopically large systems, are easily observed in the magnetization of a macroscopic sample. Experimental[6–9] as well as theoretical[10,11] work in this area has focused on the tunneling between magnetization states in magnetic macromolecules in crystals of manganese acetate (Mn_{12}ac) (Refs. 6 and 8) and iron (Fe_8) (Refs. 7 and 9) at temperatures of the order of tenths of Kelvins. At these temperatures, it is often possible to observe coherent quantum effects with minimal environment-induced dephasing and relaxation. An essential feature of these materials has been their ''giant spin'' nature; Mn_{12}ac and Fe_8 have spin quantum numbers of 10 and 8, respectively. Low-temperature studies on dilute systems such as ^{133}Cs have been carried out using state-selective Rabi and Ramsey magnetic resonance[12] and it has been demonstrated that it is possible to prepare arbitrary superposition states of the Zeeman sublevels.[13]

In this paper, we study a collection of noninteracting spins coherently driven by crossed time-dependent magnetic fields and predict an interesting resonance phenomenon that should appear in these systems. The Hamiltonian is

$$H=\Delta_0 f(t)\hat{J}_z+\Delta_x g(t)\hat{J}_x+\Delta_y h(t)\hat{J}_y, \tag{1}$$

where $\Delta_0=g\mu_B B_z, \Delta_x=g\mu_B B_x$, and $\Delta_y=g\mu_B B_y$, g is Lande's factor, and μ_B is Bohr's magneton. Here, and throughout the paper, $\hbar$ has been set to 1. The time dependences of the *x*-, *y*-, and *z*-directional fields are denoted by $g(t), h(t)$, and $f(t)$, respectively. The operators $\hat{J}_i(i=x,y,z)$ are the standard angular momentum (spin) operators for a system of total spin *j*. Denoting by *m* the quantum number for the *z* projection of the spin, we can write

$$J_\pm|j,m\rangle=(J_x\pm iJ_y)|j,m\rangle=\sqrt{(j\mp m)(j\pm m+1)}|j,m\pm 1\rangle \tag{2}$$

and express an arbitrary state as

$$|\psi(t)\rangle=\sum_{m=-j}^{m=j}C_m(t)|j,m\rangle. \tag{3}$$

Given Eqs. (1) and (2), the equation of motion for the amplitudes $C_m(t)$ is

$$\begin{aligned} i\dot{C}_m(t)=&\Delta_0 f(t)mC_m(t)+\frac{1}{2}[\Delta_x h(t)-i\Delta_y g(t)]\\ &\times\sqrt{(j+m)(j-m+1)}C_{m-1}(t)\\ &+\frac{1}{2}[\Delta_x h(t)+i\Delta_y g(t)]\\ &\times\sqrt{(j-m)(j+m+1)}C_{m+1}(t). \end{aligned} \tag{4}$$

Resonance effects. In order to represent commonly encountered magnetic macromolecules such as Mn_{12}ac and Fe_8, we take our system to have a total spin of $j=10$ and solve Eq. (4) numerically for initial occupation of the state $m=0$. Striking resonance effects emerge and are displayed in Fig. 1, in which the occupation probability of the initially occupied level is plotted as a function of time. The time dependence of the magnetic field in the *z* direction is taken throughout this paper to be

$$f(t)=\cos\omega_0 t. \tag{5}$$

The time dependence of the fields in the transverse (*x* and *y*) directions is taken to be $g(t)=h(t)=\sin\omega_0 t$ in Fig. 1(a) and $g(t)=h(t)=\cos 2\omega_0 t$ in Fig. 1(b). The probability of the initially occupied level executes oscillations such that the average value remains close to unity when the ratio of the longitudinal magnetic field energy to the longitudinal field frequency (Δ_0/ω_0) satisfies a certain resonance condition. When the condition is not satisfied, the initially occupied

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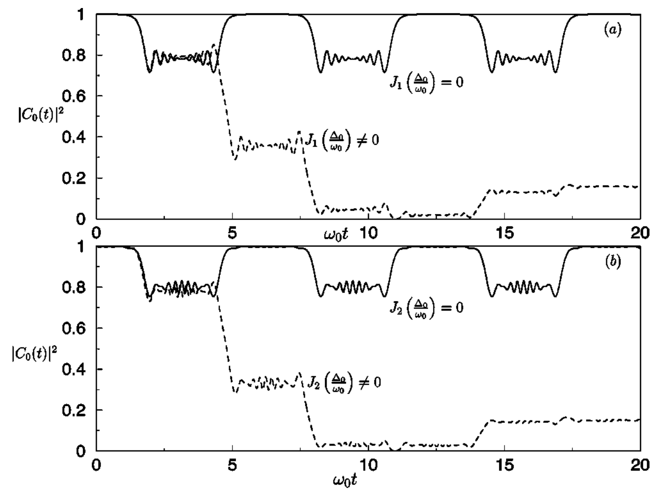

FIG. 1. The probability of the initially occupied level $|C_0(t)|^2$ as a function of dimensionless time $\omega_0 t$. The time dependence of the transverse fields is (a) $g(t)=h(t)=\sin\omega_0 t$, (b) $g(t)=h(t)=\cos 2\omega_0 t$. In both (a) and (b), the strengths of the transverse fields are taken to be $B_x=B_y=0.1B_z$. In (a), the solid and dotted lines are plotted using $\Delta_0=29.05\omega_0$ and $\Delta_0=30.62\omega_0$, respectively. In (b), the solid and dotted lines are plotted using $\Delta_0=27.42\omega_0$ and $\Delta_0=28.99\omega_0$ respectively.

level becomes depopulated rapidly. The behavior is highly reminiscent of the dynamic localization reported by Dunlap and Kenkre[14] in the context of a charge moving in a crystal,[15] as well as of trapping in two-level atoms,[16] and the work of Raghavan *et al.* in the context of driven transport in finite chains.[17]

The resonance condition in both cases shown above involves the root of Bessel functions. In Fig. 1(a), in which the transverse time dependence is $\sin\omega_0 t$, the root is of the Bessel function of the order 1, whereas in Fig. 1(b), in which the tranverse time dependence is $\cos 2\omega_0 t$, the root is of the Bessel function of order 2. Indeed, we find by numerical analysis for a variety of transverse driving fields that the following resonance conditions for localization apply when the longitudinal field dependence is given by Eq. (5):

$$g(t)=h(t)=\cos(2n\omega_0 t),\quad J_{2n}\left(\frac{\Delta_0}{\omega_0}\right)=0;\quad n=0,1,2,\ldots, \tag{6}$$

$$g(t)=h(t)=\sin[(2n+1)\omega_0 t],\quad J_{2n+1}\left(\frac{\Delta_0}{\omega_0}\right)=0;$$

$$n=0,1,2,\ldots. \tag{7}$$

These resonance conditions appear to be intimately related to, and to represent generalizations of, the Bessel root condition for dynamic localization.[14] In order to check this connection, we consider the case of constant transverse fields which reduces Eq. (4) to

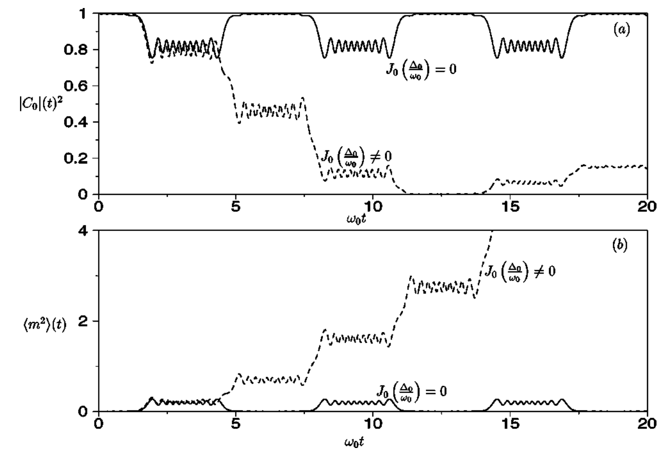

FIG. 2. (a) $|C_0(t)|^2$ as a function of $\omega_0 t$. (b) The mean-square displacement in m space as a function of $\omega_0 t$. In both (a) and (b), the transverse fields are constant, their strengths are taken to be $B_x=B_y=0.1B_z$, and the solid and dotted lines are plotted using $\Delta_0=30.64\omega_0$, and $\Delta_0=29.06\omega_0$, respectively.

$$\begin{aligned} i\dot{C}_m(t)&=\Delta_0\cos(\omega_0 t)mC_m(t)\\ &\quad+\frac{1}{2}(\Delta_x-i\Delta_y)\sqrt{(j+m)(j-m+1)}C_{m-1}(t)\\ &\quad+\frac{1}{2}(\Delta_x+i\Delta_y)\sqrt{(j-m)(j+m+1)}C_{m+1}(t). \end{aligned} \tag{8}$$

The numerical solution of Eq. (8) enables us to plot in Fig. 2(a), the occupation probability of the initially occupied level, and in Fig. 2(b), a measure of the localization of the system, the ''mean-squared displacement'' defined as $\langle m^2\rangle(t)=\Sigma_m m^2|C_m(t)|^2$. We find precisely the behavior reported in the dynamic localization study,[14] wherein a charged particle subjected to an oscillating electric field in a crystal can be localized when certain resonance conditions involving Bessel functions are satisfied. If we initially populate a state (or sublevel) m such that $|m|\ll j$ and $j\gg 1$, we can write down Eq. (8) approximately as

$$\begin{aligned} i\dot{C}_m&=\Delta_0\cos(\omega_0 t)mC_m+\frac{\Omega}{2}(C_{m+1}+C_{m-1})\\ &\quad+\frac{i\Delta_1}{2}(C_{m+1}-C_{m-1}), \end{aligned} \tag{9}$$

where $\Delta_x j=\Omega,\Delta_y j=\Delta_1$. Of course, Eq. (9) is nothing but the finite-chain equivalent of a charge moving in a finite lattice with a time-dependent, complex nearest-neighbor matrix element. We find, further, that the essential physics of the system can be described well by Eq. (9) provided that the strengths of the transverse fields are small compared to the longitudinal field, i.e., $\Omega j,\Delta_1 j\ll\Delta_0$. This is borne out by our numerical work for a wide range of parameter values of interest.

Analytic approximation procedure. We are primarily concerned with localization effects and are dealing with the regime where the transverse field strengths are weak compared to the longitudinal field strength. Therefore, we can assume that the initially localized system does not feel the ‘‘ends’’ of the spin ladder and can take, for calculational purposes, the extension of the spin ladder to be infinite. Discrete Fourier transforms are therefore useful. Following the procedure of Ref. 14, we define $C^k=\Sigma_{m=-\infty}^{m=\infty}C_m e^{ikm}$, write Eq. (9) in ‘‘reciprocal space,’’

$$\frac{\partial C^k}{\partial t}+\Delta_0 f(t)\frac{\partial C^k}{\partial k}=-i[\Omega g(t)\cos k-\Delta_1 h(t)\sin k]C^k, \tag{10}$$

apply the method of characteristics,[14,18] and invert back to m space, to obtain the amplitude in spin space:

$$C_m(t)=\sum_r C_r(0)\exp\left\{ir\Delta\int_0^t f(t')dt'\right\}\lambda^{(r-m)}J_{r-m}\times[\sqrt{\mathcal{A}(t)^2+\mathcal{B}(t)^2}], \tag{11}$$

$$\lambda=\frac{\mathcal{A}(t)-i\mathcal{B}(t)}{\sqrt{\mathcal{A}(t)^2+\mathcal{B}(t)^2}}, \tag{12}$$

$$\mathcal{A}(t)=\Omega\int_0^t dt' g(t')\sin\chi(t',t)+\Delta_1\int_0^t dt' h(t')\cos\chi(t',t), \tag{13}$$

$$\mathcal{B}(t)=\Omega\int_0^t dt' g(t')\cos\chi(t',t)-\Delta_1\int_0^t dt' h(t')\sin\chi(t',t), \tag{14}$$

$$\chi(t',t)=\Delta_0\int_t^{t'} ds f(s). \tag{15}$$

If we are interested in the initial population being in spin-sublevel 0, we have $C_r(0)=\delta_{r,0}$, and obtain from Eq. (11),

$$C_m(t)=\lambda^{-m}J_{-m}[\sqrt{\mathcal{A}(t)^2+\mathcal{B}(t)^2}], \tag{16}$$

for the amplitudes, and

$$|C_m(t)|^2=J_m^2[\sqrt{\mathcal{A}(t)^2+\mathcal{B}(t)^2}] \tag{17}$$

for the probabilities. The expression for the mean-squared displacement reads

$$\langle m^2\rangle(t)=\frac{1}{2}[\mathcal{A}(t)^2+\mathcal{B}(t)^2]. \tag{18}$$

For transverse field dependences such that $g(t)=h(t)$, we can simplify many of the relations obtained above to obtain

$$|C_m(t)|^2=J_m^2[\sqrt{\mathcal{A}(t)^2+\mathcal{B}(t)^2}]=J_m^2(\{(\Omega^2+\Delta_1^2)[u(t)^2+v(t)^2]\}^{1/2}), \tag{19}$$

$$\langle m^2\rangle(t)=\frac{1}{2}\{(\Omega^2+\Delta_1^2)[u(t)^2+v(t)^2]\}, \tag{20}$$

where

$$u(t)=\int_0^t dt' h(t')\cos\left[\Delta_0\int_0^{t'} ds f(s)\right],$$

$$v(t)=\int_0^t dt' h(t')\sin\left[\Delta_0\int_0^{t'} ds f(s)\right]. \tag{21}$$

We thus see that the localization condition is obtained from the boundedness (or lack thereof) of the functions $u(t)$ and $v(t)$.

When the transverse field dependence is $g(t)=h(t)=\cos(2n\omega_0 t)$, $n=0,1,2,\ldots,$

$$\begin{aligned}u(t)&=\frac{1}{\omega_0}\int_0^{\omega_0 t}d\tau\cos 2n\tau\cos\left[\frac{\Delta_0}{\omega_0}\sin\tau\right]\\&=J_{2n}\left(\frac{\Delta_0}{\omega_0}\right)t+\frac{1}{\omega_0}\left[J_0\left(\frac{\Delta_0}{\omega_0}\right)\frac{\sin 2n\omega_0 t}{2n}+\sum_{k=1,k\neq n}^{\infty}J_{2k}\left(\frac{\Delta_0}{\omega_0}\right)\right.\\&\quad\left.\times\left(\frac{\sin 2(n+k)\omega_0 t}{2(n+k)}+\frac{\sin 2(n-k)\omega_0 t}{2(n-k)}\right)\right],\end{aligned} \tag{22}$$

$$\begin{aligned}v(t)&=\frac{1}{\omega_0}\int_0^{\omega_0 t}d\tau\cos 2n\tau\sin\left[\frac{\Delta_0}{\omega_0}\sin\tau\right]\\&=\frac{1}{\omega_0}\sum_{k=1}^{\infty}J_{2k-1}\left(\frac{\Delta_0}{\omega_0}\right)\left[\frac{1-\cos(2n+2k-1)\omega_0 t}{2n+2k-1}\right.\\&\quad\left.+\frac{1-\cos(2k-1-2n)\omega_0 t}{2k-1-2n}\right].\end{aligned} \tag{23}$$

The function $u(t)$ is bounded only if $J_{2n}(\Delta_0/\omega_0)=0$, whereas $v(t)$ is always bounded. Hence the system is localized dynamically whenever $J_{2n}(\Delta_0/\omega_0)=0$. Note, as a particular case, that when the transverse fields are constant, $n=0$ above, and the Bessel function of order 0 appears, as in the work of Dunlap and Kenkre.[14]

When the transverse field dependence is $g(t)=h(t)=\sin(2n-1)\omega_0 t, n=1,2,\ldots,$ we obtain

$$\begin{aligned}u(t)&=\frac{1}{\omega_0}\int_0^{\omega_0 t}d\tau\sin(2n-1)\tau\cos\left[\frac{\Delta_0}{\omega_0}\sin\tau\right]\\&\quad+\sum_{k=1}^{\infty}J_{2k}\left(\frac{\Delta_0}{\omega_0}\right)\left(\frac{1-\cos(2n+2k-1)\omega_0 t}{2n+2k-1}\right.\\&\quad\left.+\frac{1-\cos(2n-2k-1)\omega_0 t}{2n-2k-1}\right),\end{aligned} \tag{24}$$

which is always bounded. For $v(t)$, we have

$$\begin{aligned}v(t)&=\frac{1}{\omega_0}\int_0^{\omega_0 t}d\tau\sin(2n-1)\tau\sin\left[\frac{\Delta_0}{\omega_0}\sin\tau\right]\\&=\frac{1}{\omega_0}\sum_{k=1,k\neq n}^{\infty}J_{2k-1}\left(\frac{\Delta_0}{\omega_0}\right)\left[\frac{\sin 2(n-k)\omega_0 t}{2(n-k)}\right.\\&\quad\left.-\frac{\sin 2(n+k-1)\omega_0 t}{2(n+k-1)}\right],\end{aligned} \tag{25}$$

which is bounded only if $J_{2n-1}(\Delta_0/\omega_0)=0$.

We thus see that the simple approximation procedure based on the replacement of the exact Eq. (4) by Eq. (9), in which the square-root terms are replaced by constants and the extension in m space is taken infinite, reproduces the resonance conditions (7).

It is not surprising that resonance effects of the kind we have studied above occur for driven magnetic systems. What is of special interest is that great control over the effects is possible by a manipulation of the time dependence of the three independent magnetic fields. Of the multitude of possibilities, we have chosen for illustration a family of cases wherein the transverse fields are equal to each other and all fields are sinusoidal. We have found that the relationship of the frequencies and phases of the transverse fields to those of the longitudinal field dictates what order of Bessel functions is involved in the resonance condition. The complicated matrix elements characteristic of angular momentum operators [see Eq. (4)] result, for the time and parameter ranges of interest, in behavior well approximated by simple equations that are translationally invariant in m space.

The translationally invariant Eq. (9), while an approximation for the magnetic systems we have considered, is in fact an exact starting point for the analysis of the original dynamic localization effect[14,15,19] if the intersite transfer integrals (bandwidths) are time-dependent and complex. Such a situation is difficult to arrange in the context of charges moving in a crystal under the action of an electric field. However, it appears possible, and indeed, easily manageable, in optical lattices.[19] The treatment we have presented above in the form of an analytic procedure is thus, applicable for optical lattices without approximation. We hope that such experiments will be undertaken. Other experimental manifestations of the resonance phenomenon in spin systems would be apparent in observations of the spin-correlation function and the scattering function of probe particles such as neutrons. While a detailed analysis of such suggested experiments will be given elsewhere, we mention in passing that the zero-frequency component of the van Hove scattering function for these systems turns out to be inversely proportional to the square root of $J_{2n}(\Delta_0/\omega_0)$, and $J_{2n+1}(\Delta_0/\omega_0)$, respectively, for the two cases where the transverse fields are $g(t)=h(t)=\cos(2n)\omega_0 t$ and $g(t)=h(t)=\sin(2n+1)\omega_0 t$. It is clear that the singularity that the scattering function develops at zero frequency is representative of dynamic localization.

This work was supported in part by the National Science Foundation under Grant Nos. DMR-9614848 and PHY 94-15583. Work at Los Alamos was performed under the auspices of the USDOE.

[1] J. L. V. Hemmen and A. Sütö, Physica B & C **141B**, 37 (1986).
[2] M. Enz and R. Schilling, J. Phys. C **19**, 1765 (1986).
[3] E. M. Chudnovsky and L. Gunther, Phys. Rev. Lett. **60**, 661 (1998).
[4] P. C. E. Stamp, E. M. Chudnovsky, and L. Gunther, Int. J. Mod. Phys. B **6**, 1355 (1992).
[5] *Quantum Tunneling of the Magnetization*, Vol. 301 of *NATO Advanced Study Institute, Series E: Applied Sciences*, edited by L. Gunther and B. Barbara (Kluwer, Dordecht, 1995).
[6] L. Thomas, F. Lionti, R. Ballou, D. Gatteschi, R. Sessoli, and B. Barbara, Nature (London) **383**, 145 (1996).
[7] W. Wernsdorfer and R. Sessoli, Science **284**, 133 (1999).
[8] J. R. Friedman, M. P. Sarachik, J. Tejada, and R. Ziolo, Phys. Rev. Lett. **76**, 3830 (1996).
[9] C. Sangregorio, T. Ohm, C. Paulsen, R. Sessoli, and D. Gatteschi, Phys. Rev. Lett. **78**, 4645 (1997).
[10] V. V. Dobrovitski and A. K. Zvezdin, Europhys. Lett. **38**, 377 (1997).
[11] L. Gunther, Europhys. Lett. **39**, 1 (1997).
[12] G. Xu and D. J. Heinzen, Phys. Rev. A **59**, R922 (1999).
[13] G. Xu and D. J. Heinzen (unpublished).
[14] D. H. Dunlap and V. M. Kenkre, Phys. Rev. B **34**, 3625 (1986); **37**, 6622 (1988); Phys. Lett. A **127**, 438 (1988).
[15] M. Holthaus, Phys. Rev. Lett. **69**, 351 (1992); D. Cai, A. R. Bishop, and N. Gronbech-Jensen, *ibid.* **72**, 591 (1994).
[16] G. S. Agarwal and W. Harshawardhan, Phys. Rev. A **50**, R4465 (1994).
[17] S. Raghavan, V. M. Kenkre, D. H. Dunlap, A. R. Bishop, and M. I. Salkola, Phys. Rev. A **54**, R1781 (1996) (see, in particular, Fig. 4 of this reference).
[18] A. N. Tikhonov, B. Vasil'eva, and A. G. Sveshnikov, *Differential Equations* (Springer, Berlin, 1985).
[19] K. W. Madison, M. C. Fischer, R. B. Diener, Q. Niu, and M. G. Raizen, Phys. Rev. Lett. **81**, 5093 (1998).