Nonlinear spin excitations in finite Heisenberg chains

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In the recent literature, there have been discussions of nonlinear spin excitations in one-dimensional, anisotropic Heisenberg spin chains. These have the character of localized excitations, which emerge from the time-dependent classical equations of motion. We show that for a given frequency, in a finite spin chain one has a hierarchy of nonlinear excitations, whose envelopes have the appearance of a one soliton, two soliton, . . . , states. Also, we consider the nature of these nonlinear excitations in an applied magnetic field of frequency ω, and explore aspects of the transient behavior of the system in response to a time varying external field. [S0163-1829(96)05437-9]

I. INTRODUCTION

The elementary excitations out of the ground state of a Heisenberg magnet have been understood for many decades. These are spin waves[1] and at low temperatures the thermal excitation of low lying spin waves provides the dominant contribution to those thermodynamic properties influenced by the magnetic degrees of freedom.

It was argued some years ago[2] that in one-dimensional spin chains, solitons also appear as elementary excitations. Before this was noted, in three-dimensional materials, a description of the classical Bloch wall in a ferromagnet emerges from the static soliton solution of the well known Sine-Gordon equation, formed here by seeking spin configurations which render the energy an extremum.[3] In three-dimensional crystals, the area of such a Bloch wall is macroscopic, with the consequence its excitation energy is macroscopic, and is hence very large compared to k_BT. Thus, these entities make no contribution to the thermodynamics of the material. But for a suitable one-dimensional spin chain, one may have solitons whose excitation energy is comparable to k_BT, simply because their cross-sectional area is microscopic.

The solitons just described, when at rest, emerge as *time-independent* solutions of the equations of motion. The solitons may also move with some velocity;[4] thus in one-dimensional spin chains, at suitably low temperature, one may view the thermal excitations as spin waves, supplemented by a dilute gas of thermally excited magnetic solitons.

Recently, in the theoretical literature,[5] a new nonlinear spin excitation in one-dimensional spin systems has been studied. These have envelope functions with a shape familiar from the theory of solitonlike objects, but they emerge as solutions of the *time-dependent* classical equations of motion. When such an entity is present, each spin in the system engages in circular precession, with a frequency Ω that lies outside the spin wave band of linear theory. These states are magnetic analogues of the intrinsic anharmonic localized modes discussed very actively in literature on the vibrations of one-dimensional anharmonic chains.[6] At the time of this writing, the means of exciting the new nonlinear magnetic excitations, or their possible role in the thermodynamics, is unclear.

In this paper, we examine the nature of such modes in finite spin chains. It should be remarked that we have in mind the possibility that these entities may exist in magnetic superlattices, some of which are described by an energy functional quite identical to that used earlier,[5] and in the present paper. The role of the single spin in the present paper is played by the total magnetic moment of a ferromagnetic film in a superlattice. Such superlattices are in fact a physical realization of a finite spin chain. The system of ''spins'' is truly classical here in that the total spin moment of each film is macroscopic. We shall elaborate on these comments below.

In the spin chain of infinite length, the intrinsic localized spin modes have an envelope function that has the character of a single soliton.[7] We find here a hierarchy of solutions associated with the finite chain. We have entities with envelopes that have the appearance of one soliton, two soliton, three soliton, . . . , states. We also show that if a circular polarized field of frequency ω is applied perpendicular to the Zeeman field, we still have such hierarchy of states and we outline their properties. Finally, we inquire if a transient, spatially uniform field can excite these states. We find the answer is negative for the scheme investigated.

II. NONLINEAR SPIN EXCITATIONS IN FINITE CHAINS: BASIC PROPERTIES

We consider a finite chain of N spins, described by the Hamiltonian

$$H=-2J\sum \mathbf{S}_n\cdot\mathbf{S}_{n+1}+A\sum (S_n^z)^2-H_0\sum S_n^z$$
$$-\sum h_n(S_n^x\cos\omega t-S_n^y\sin\omega t). \qquad (1)$$

The spins are ferromagnetically coupled through nearest-neighbor exchange interaction J. We have single site anisotropy which renders the xy plane an easy plane. An external field H_0 is applied along the z direction. We shall assume H_0 is large enough that in the ground state, the spins are parallel to H_0 and the z axis, and thus are perpendicular to the easy plane. This requires $H_0>AS$. Furthermore, we assume a circularly polarized field of frequency ω is applied in the xy

plane. The strength of the field on site n is h_n. This is the Hamiltonian which formed the basis for a discussion of nonlinear spin excitations in the infinite chain if we set $h_n=0$[5].

As noted earlier, the spin Hamiltonian in Eq. (1) is applicable to the quasi-one-dimensional material $CsFeCl_3$.[8] A rather large applied field, in the range of 40 T will be required to align the spins parallel to the z axis. There are also a number of magnetic superlattice structures described by the energy functional in Eq. (1). Consider a superlattice fabricated from very thin ferromagnetic Fe films, separated by nonferromagnetic metal spacers. It is well known that magnetic couplings of exchange character between the Fe films are transmitted through the spacer layer. These oscillate in sign as the thickness of the spacer layer is varied[9] and thus may be arranged to be ferromagnetic. The Fe films often have magnetization in plane with strong anisotropy of uniaxial character. One may move the moments out of the plane and align them perpendicular to it through application of modest external fields H_0, for suitable samples. We may apply the Hamiltonian in Eq. (1) directly to such materials, provided we associate $\mathbf{S}_i$ with the *total* spin moment of a particular Fe film, rather than that of a single atom, as one does in the discussion of quasi-one-dimensional magnetic materials. Such magnetic superlattices also have a finite number of layers, and thus are a realization of a finite spin chain. Notice that the dynamics of such a structure is described quite precisely by the *classical*, rather than the quantum mechanical, equations of motion, since here the spin $\mathbf{S}_i$ is very large.

We consider, as before,[5] the equation of motion for the operator $S_n^+=S_n^x+iS_n^y$. We have

$$i\hbar\,\frac{\partial S_n^+}{\partial t}=H_0S_n^+ +2J[S_n^+(S_{n+1}^z+S_{n-1}^z)-S_n^z(S_{n+1}^+ +S_{n-1}^+)]$$
$$-2AS_n^zS_n^+ -h_ne^{-i\omega t}S_n^z. \tag{2}$$

We have, for these classical spins,

$$S_n^z=\sqrt{S^2-S_n^+S_n^-}=\sqrt{S^2-S_n^+(S_n^+)}. \tag{3}$$

The equation of motion admits time-dependent solutions which we write, with s_n real,

$$S_n^+=Ss_ne^{-i\omega t}, \tag{4}$$

where

$$S_n^z=S\sqrt{1-(s_n)^2} \tag{5}$$

is in fact independent of time. Thus, all spins are engaging in circular precession on a cone which makes the angle $\theta_n=\sin^{-1}(s_n)$ with the z axis.

We have the following time independent equation for the amplitude s_n:

$$\Omega s_n=s_n(\sqrt{1-s_{n+1}^2}+\sqrt{1-s_{n-1}^2})-(s_{n+1}+s_{n-1})\sqrt{1-s_n^2}$$
$$-2Bs_n\sqrt{1-s_n^2}-c_n\sqrt{1-s_n^2}, \tag{6}$$

where

$$\Omega=\frac{(\omega-H_0)}{2JS}, \tag{7}$$

$$B=\frac{A}{2J}, \tag{8}$$

and

$$c_n=\frac{h_n}{2JS}. \tag{9}$$

If we set c_n to zero, and consider small amplitude solutions, we have the standard spin waves. For these, $s_n=s\exp(ikn)$, and the spin wave dispersion relation has the form, in the terms of the dimensionless variables in Eq. (6)

$$\Omega(k)=-2B+2(1-\cos k). \tag{10}$$

The spin wave bands thus occupy the frequency domain $-2B\leqslant\Omega\leqslant 4-2B$.

We have free ends on our finite chain, and the equations of motion for the end spins differ from Eq. (6). We call the two end spins $n=1$, and $n=N$. For the spin $n=1$ we have

$$\Omega s_1=s_1\sqrt{1-s_2^2}-s_2\sqrt{1-s_1^2}-2Bs_1\sqrt{1-s_1^2}-c_1\sqrt{1-s_1^2}. \tag{11}$$

A similar equation applies to the other end of the chain, $n=N$.

The intrinsic localized spin excitations studied here occur for frequencies which lie above the linear spin wave bands. We discuss the various examples we have explored. We proceed by solving the above system of equations on a computer.

In the numerical calculations, we proceed as follows. We guess for s_1 and use Eq. (11) to solve for s_2. Then given s_1 and s_2, we may generate s_3 from Eq. (6). In this manner we proceed through to the end of the chain and interrogate the equation of motion for spin $n=N$ to see if it is satisfied. We generate solutions by scanning the initial value of s_1.

We now turn to the various cases we have explored.

A. Multi-soliton states in zero driving field

We begin with results for the homogeneous, nonlinear equation of motion formed by setting $h_n=c_n=0$ for all spins. It is convenient to write, in some instances,

$$s_n=(-1)^nf_n, \tag{12}$$

where for frequencies Ω above the linear spin wave bands, we find f_n to be a slowly varying envelope function. We have selected $B=4$. The linear spin wave bands then extend from $\Omega=-8$ to $\Omega=-4$, in our units. The numerical calculations reported here employ $\Omega=-3.95$, so we have a frequency a bit above the linear spin wave band.

In Fig. 1, we show two examples of a one soliton state, for a line of 101 spins. Here we display s_n. There are two distinct solutions of different symmetry. For the first, s_n is odd under reflection through the midpoint of the line and for the second, s_n is even under this reflection. For the first case, one has $s_1=s_N=1.345\times10^{-6}$ and for the second, one has $s_1=s_N=1.203\times10^{-6}$. While we show two distinct solutions in Fig. 1, each of these states is in fact twofold degenerate. This is because Eq. (6) is left invariant under the transforma-

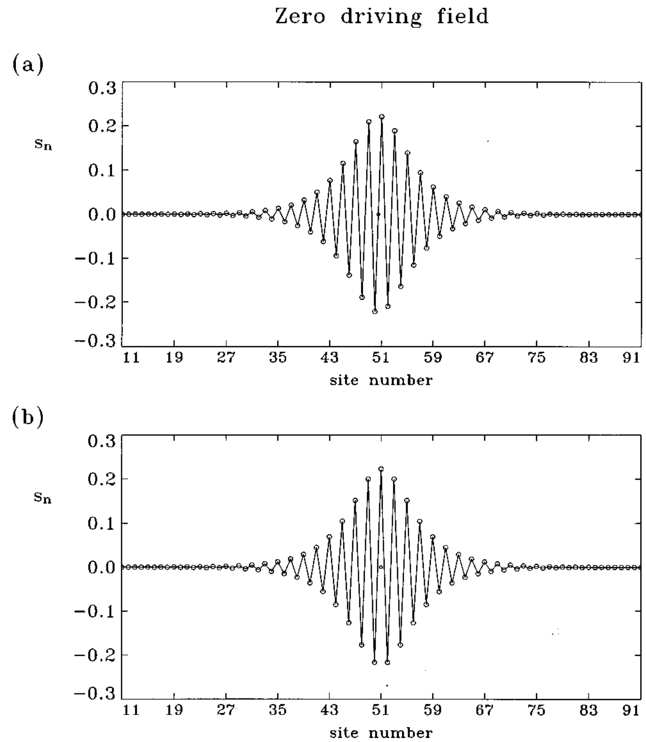

FIG. 1. We show the function s_n for two one soliton states associated with a chain of 101 spins. We have (a) an odd parity state and (b) an even parity state. The calculations employ $\Omega = -3.95$ and we have taken $B=4$. The externally applied field $h_n=0$. For case (a), $s_1=s_N=1.345\times10^{-6}$, while for (b) we have $s_1=s_N=1.203\times10^{-6}$.

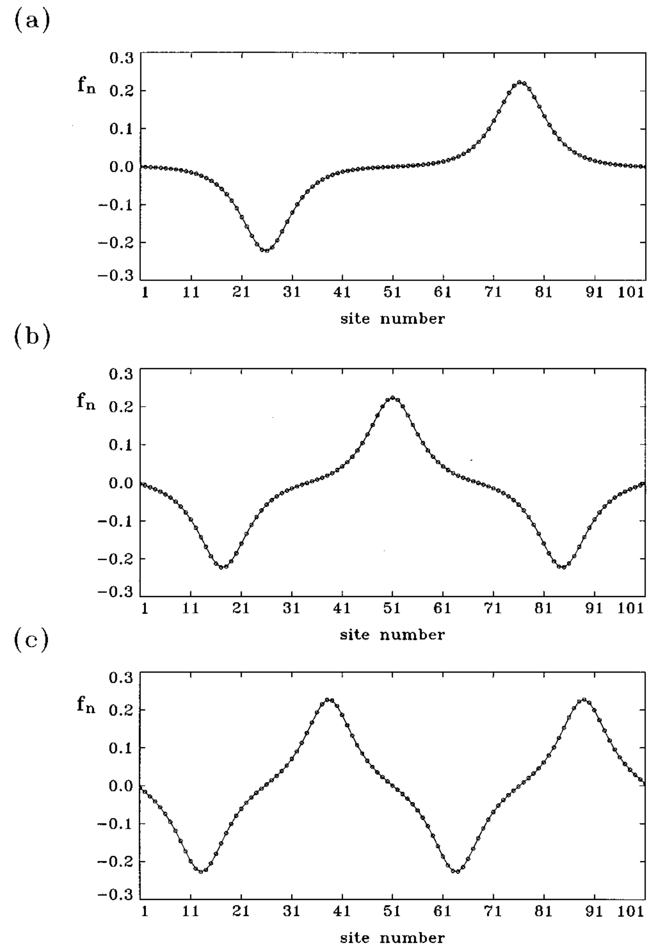

FIG. 2. For the frequency used in Fig. 1 and also $B=4$, we show the envelope function f_n for (a) a two soliton state, (b) a three soliton state, and (c) a four soliton state. For (a) we have $s_1=s_N=3.365\times10^{-4}$, for (b) $s_1=s_N=2.177\times10^{-3}$, and for (c) $s_1=s_N=5.000\times10^{-3}$.

tion $s_n \rightarrow -s_n$, so long as $c=0$. Notice this symmetry does not hold in a driving field when $c\neq0$.

As stated above, for the same frequency Ω used in Fig. 1, we have a whole hierarchy of soliton states. We show examples in Fig. 2, for exactly the same frequency Ω employed in Fig. 1. Here we display the envelope functions f_n, rather than s_n.

Thus, for the finite chain and a given value of the frequency Ω, we find a hierarchy of multisoliton states, as remarked earlier. So far as we know, we could generate states with five, six, or more solitons, though we have not explored how far we can proceed with the hierarchy. Such a hierarchy will exist for a finite chain to which periodic boundary conditions are applied as well, though this sums to be a case of primarily academic interest. For the infinitely long chain, and the boundary conditions $s_n \rightarrow 0$ as $n\rightarrow\pm\infty$, we believe only the even and odd parity one soliton states are present.

We have explored the stability of the states described here and found them quite stable against small amplitude perturbations. This has been done as follows. We imagine that at some time, $t=0$, the spins are frozen with a pattern like that displayed in, say, Fig. 1(a) or Fig. 1(b). We perturb this pattern by adding small increments δs_n to the various spin deviations. We use the pattern so generated as an initial condition for Eq. (2). We study numerical solutions after inserting the form $s_n^+ = S s_n(t) e^{-i\omega t}$ into these equations, where now we allow $s_n(t)$ to be complex. [When $s_n(t)$ is not real, then in Eq. (6) and elsewhere, $\sqrt{1-s_n^2}$ and $\sqrt{1-s_{n\pm1}^2}$ are replaced by $\sqrt{1-|s_n|^2}$, etc.] We find $s_n(t)$ executes stable small amplitude oscillations around the soliton states. One may describe the subsequent behavior of the system by saying that the soliton ''pulses'' execute stable periodic oscillations in amplitude. We see no evidence that the solitons break up into spin waves, for example. The energy density remains always concentrated in the solitonic peaks.

This situation is most intriguing, since we have a complex hierarchy of states associated with any frequency. There is nothing special about our choice $\Omega=-3.95$, as far as we can see. We can generate similar hierarchies for any frequency above the maximum linear spin wave frequency.

B. Nonlinear spin excitations in the presence of a spatially uniform driving field

We next turn to the response of the system when a spatially uniform, circularly polarized field is present, which oscillates with frequency ω. We include its presence by choosing h_n in Eq. (2) equal to h, independent of the site index n. We are then able to solve Eq. (6) in combination with Eq. (11) for $c_n=c$ independent of n.

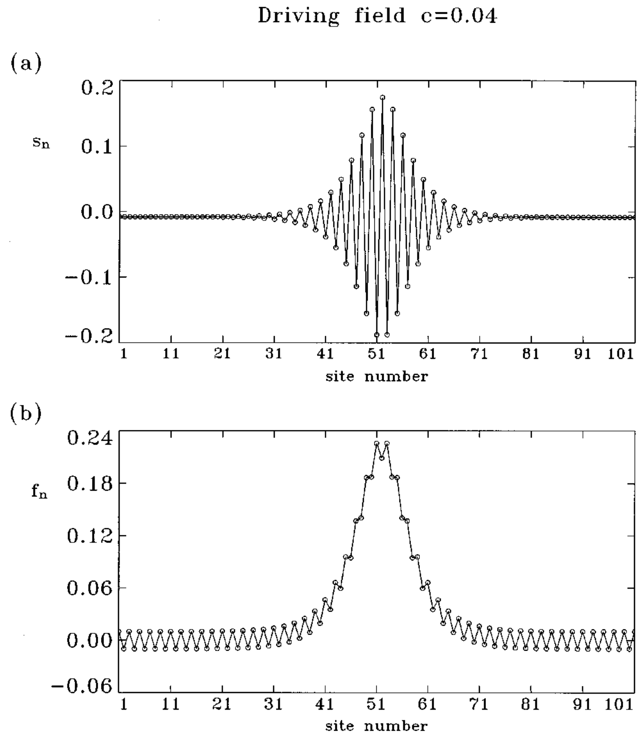

FIG. 3. We show the functions (a) s_n and (b) $f_n = (-1)^n s_n$ for a nonlinear spin excitation in the presence of in rf field. Here the dimensionless field strength $c = 0.04$ and the remaining parameters are the same as in Figs. 1 and 2.

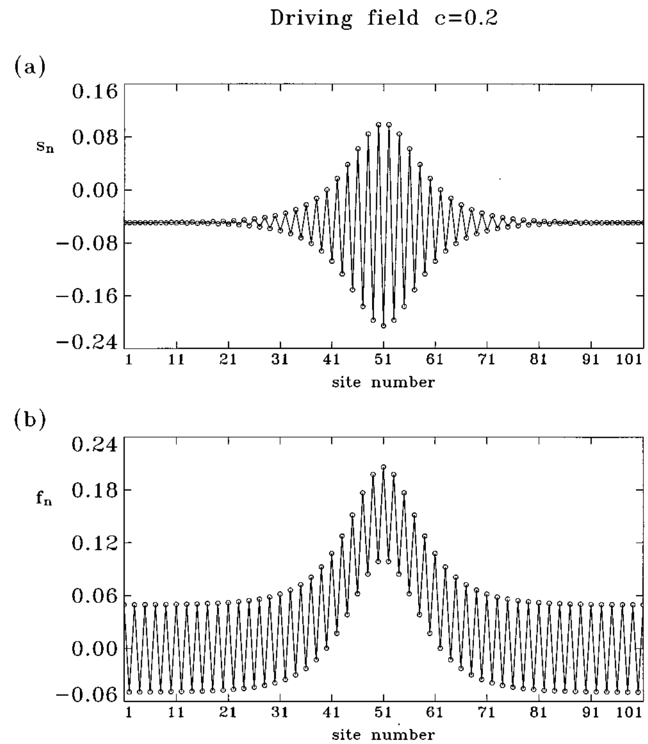

FIG. 4. The same as Fig. 3 but now $c = 0.20$.

For any choice of c we always have a uniform solution in which s_n is independent of n. For this uniform precession of all the spins the exchange couplings drop out and the equations for both interior and end spins degenerate into

$$\Omega s_0 = -2Bs_0\sqrt{1-s_0^2} - c\sqrt{1-s_0^2}. \tag{13}$$

If the amplitude of the driving field c is very small, linearization of Eq. (13) gives

$$s_0 = -\frac{c}{(2B+\Omega)} \equiv \frac{c}{(\Omega_m - \Omega)}, \tag{14}$$

where in our units, $\Omega_m = -2B$ is the frequency of the $k=0$ spin wave mode in linear spin wave theory. Notice if $\Omega > \Omega_m$, the spins are precisely directed antiparallel to the driving field which is the behavior characteristic of a harmonic oscillator driven above its resonance frequency. As c is increased, one may follow this solution through use of Eq. (13).

Also when $c \neq 0$, we find solitonlike states in the presence of the external driving field, which, as $c \to 0$, degenerate into the structures shown in the previous section. In Fig. 3(a), for the case $c = 0.04$, we show s_n and in Fig. 3(b) we show f_n. One sees from Fig. 3(b) that the external driving field induces an oscillatory modulation in the envelope s_n. As the amplitude c becomes larger these oscillations increase in amplitude, as we see from Fig. 4.

When $c \neq 0$, the solitonic feature remains near the center of the chain, as we see from these figures. As we move out to either end of the chain the amplitude of the end spins is very close to, but distinctly different from, that associated with the uniform state found from Eq. (13).

When $c = 0$, as we noted earlier, each of the soliton states illustrated in Fig. 1 is twofold degenerate, in a certain sense. With $c = 0$, the equations are invariant under the transformation $s_n \to -s_n$ and we may form a distinct solution from, say, Fig. 1(a), by just reversing the sign of s_n everywhere.

The presence of the external field breaks this symmetry. There is a sort of ''splitting'' of the two previously degenerate solitons, as follows. For $c = 0.2$, we obtain two distinct one soliton structures with even parity. We call these the A state and the B state, respectively. For each, we illustrate the functions f_n in Fig. 5. For the A state, the value of s_1 re-

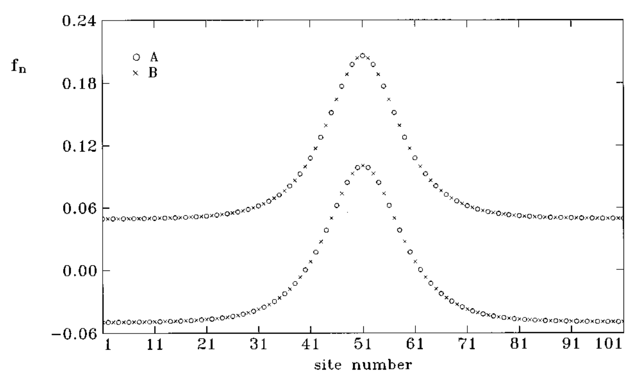

FIG. 5. For $c = 0.20$ we show a comparison between the two solitons, the type A and type B structures discussed in the text. The X's are the type A excitation, and the ○'s are the type B. We have multiplied f_n for the type A soliton by -1, for clarity of presentation. As earlier we have $\Omega = -3.95$ and $b = 4.00$.

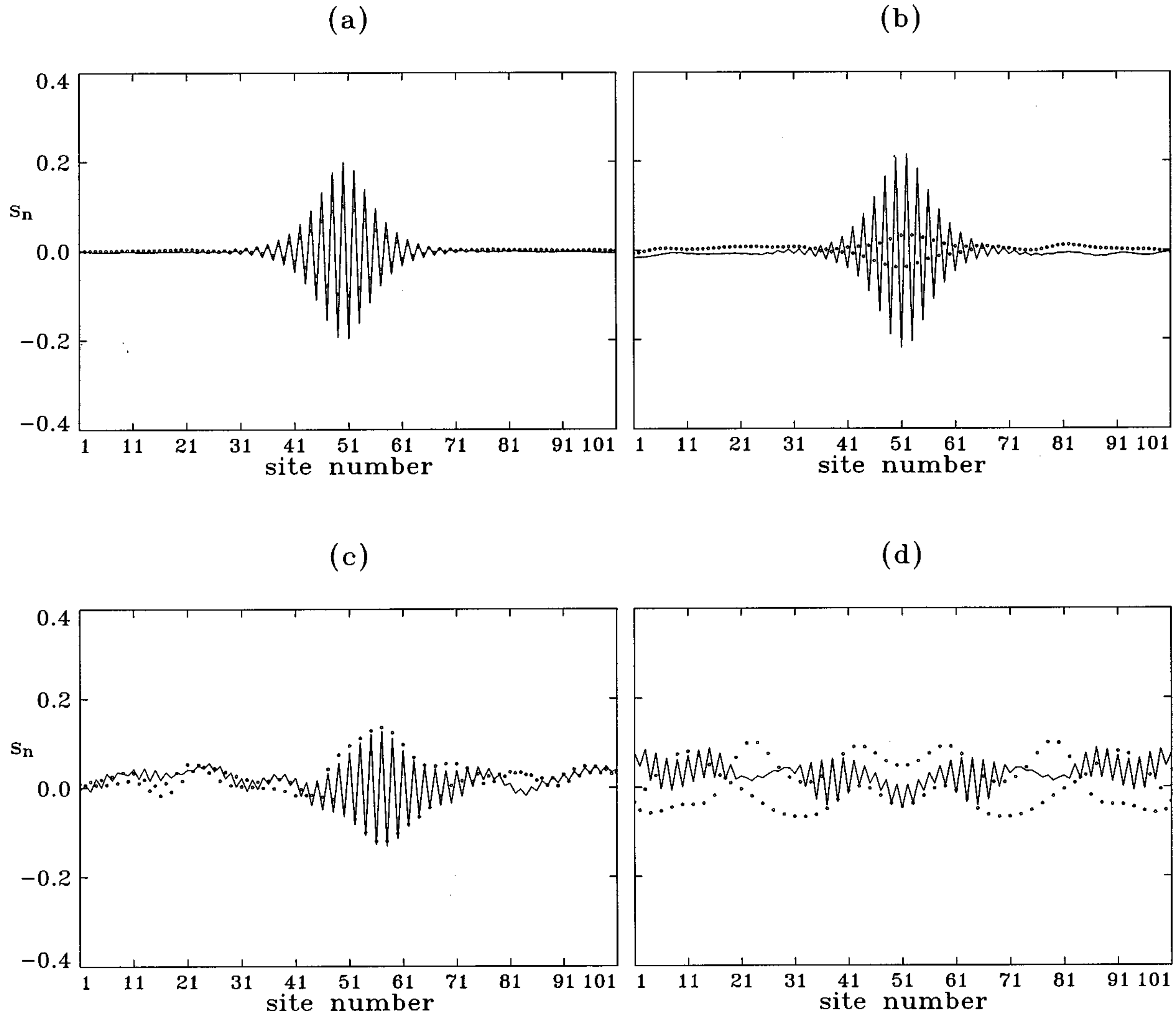

FIG. 6. An illustration of the stability of the one soliton states in the external rf field. The structure, perturbed slightly as described in the text, has been followed up to a time $t_0=500/(2JS)$. We see that in strong external fields, the lifetime of the state shortens. Each figure displays the response when the dimensionless time $t=500$. In (a), $c=0.01$, in (b), $c=0.04$, in (c), $c=0.15$, and in (d), $c=0.20$. In all cases, $\Omega=-3.95$ and $b=4.00$.

quired to generate the state is $s_1^A=-0.049\,456\,59$, while $s_1^B=-0.049\,426\,92$. The spatially uniform solution has $s_0=-0.049\,441\,76$, between these two values.

In the previous section, we discussed studies of the stability of the one soliton states, when $c=0$. As remarked there, we find these states stable over a long period of time when we use the procedure described. For $c\neq 0$ the soliton structure appears to have a finite lifetime which decreases with increasing field strength c. We illustrate this in Fig. 6. We have tested stability as follows, once again. At time $t=0$, we assign each spin a spin deviation precisely equal to that for a one soliton state, such as those illustrated in Figs. 3–5. We then add small amplitude increment δs_n to each of these spin deviations. We take, in general, δs_n to be complex. We then solve the full equation of motion through use of the ansatz $s_n^+=Ss_n(t)e^{-i\omega t}$ and we study $s_n(t)$. Our dimensionless unit of time is $(2JS)^{-1}$. We note that $2JS$ is the precession frequency of a given spin, in the exchange field provided by one of its nearest neighbors. In Fig. 6, we show the values of $s_n(t_0)$ at the time $t_0=250/JS$. This is quite a long time for the spin system, in that it corresponds to roughly 100 precessional periods in the above mentioned exchange field. In Fig. 6(a), we see $s_n(t_0)$ for a very small value of the external field, $c=0.01$. The solid line connects values of $\mathrm{Re}[s_n(t_0)]$, and the dots illustrate $\mathrm{Im}[s_n(t_0)]$. We see the localized, solitonic structure is quite stable. Once the energy density is localized at $t=0$, it remains there for quite some time. Our simulations for $c=0$ behave in a similar manner. For $c=0.04$, as we see from Fig. 6(b), we still have a central structure displayed prominently, but the soliton has clearly shed energy in the form of spin waves which propagate outward and reflect off the chain ends. By the time $c=0.15$, we see that when $t_0=250/JS$ this effect is far more pronounced and when $c=0.20$ we see no evidence left of the central feature.

These simulations thus suggest that when $c=0$, the nonlinear excitation is very long lived and stable against diverse small perturbations (we have explored several schemes for selecting δs_n). When $c \neq 0$, but not too large, it may be viewed as a long-lived nonlinear resonance structure, which decays to spin waves when perturbed slightly. Its lifetime becomes quite short when $c=0.2$. In this case, even by the time $t_0=5/JS$, we can see evidence of decay in our simulations.

In the next section we explore the possibility that suitably applied fields may excite the nonlinear resonances. Clearly, if this is to be done, the amplitude of the driving field cannot be too large, though at the same time it must be large enough to enter the domain where nonlinearities are significant.

C. Summary of a study of means of exciting intrinsic localized modes

In the above discussion, we have presented studies of aspects of intrinsic nonlinear spin excitations in a finite spin chain which, as we have argued, can serve as a model of a magnetic superlattice.

A central question, not addressed explicitly in the literature on lattice dynamics so far as we know,[6] is how one may probe or excite these modes in an experiment. At least in principle, one would like to expose the sample to an appropriate external driving field and possibly observe anomalous absorption at high power levels for frequencies above the linear spin wave band.

We have explored this possibility in two ways, each of which proved unsuccessful. In the first, we imagine the line of spins studied above is exposed to a uniform field of frequency ω, circularly polarized in xy plane, as in Eq. (1). We apply a spatially uniform field to the system, where the field felt by spin n has the form $h_n(t)e^{-i\omega t}$, with $h_n(t)=h(1-e^{-\epsilon t})$. The field thus starts at very small amplitude and builds up with time. In the early history of the time evolution linear response theory holds and a spatially uniform disturbance is induced in the spin system. The question is whether this remains stable at large final amplitudes h, or whether it becomes unstable with respect to formation of a nonlinear excitation. Our answer, based on the model used above, is that the uniform state is stable. To achieve a stable final state and damp out transients, we added phenomenological damping of the Landau-Lifshitz form $-\gamma(\mathbf{S}_n \times \dot{\mathbf{S}}_n)$ to the equation of motion of each spin. This form has the virtue that it preserves the length of each $\mathbf{S}_n$.

In the literature on one-dimensional anharmonic lattices, Kiselev and co-workers have explored the stability of various normal modes, taken to large amplitude.[10] They find the uniform is made stable at large amplitude, as we do for the spin system. We understand that Kiselev and a colleague have also explored the stability of the uniform mode in the spin system and find it stable, as we do.[11]

In the anharmonic problem, it has been demonstrated that the out-of-phase optical mode, when driven to large amplitude, is unstable and evolves into nonlinear intrinsic modes.[10] If we envision a one-dimensional line of ions, each with alternating positive and negative charge, then a spatially uniform electric field excites the out of phase mode. Unfortunately, for the spin system, a uniform field of small amplitude excites only the $k=0$ acoustic spin wave, which is the uniform mode.

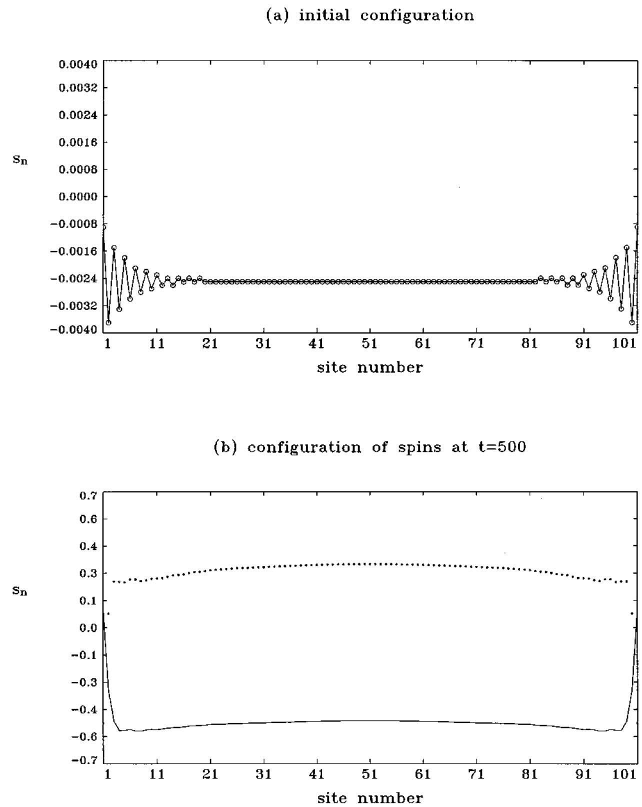

FIG. 7. (a) The response of the model spin chain to a spatially uniform driving field, as provided by linear response theory, when the anisotropy strength A for the end spins is 1.5 that of those in the bulk. (b) For the chain considered in (a) we show the response at time $t=500$, when the driving field $c=0.10$. We have added damping for each spin of the Landau-Lifshitz form so at long times a steady state is achieved.

This last statement is true only for the ideal line of exchange coupled spins, where spins located at each end of the line are described by model parameters identical to those in the interior. If we perturb the end spins, then for frequencies above the spin wave band we induce an out-of-phase disturbance near the ends of the chain. We may imagine, for example, that for the two end spins the anisotropy constant A in Eq. (1) differs from that in the interior. It will surely be the case that this is so for any finite chain of magnetic moment bearing atoms, since A is sensitive to the local site symmetry. In a superlattice, one could synthesize a structure whose end films have different anisotropy constants than the interior films. In Fig. 7, for a particular choice of A for the two end spins, we show the disturbance in the spin chain, as calculated from linear response.

By the method outlined earlier in which the external field is ramped up in strength, we have explored the stability of this disturbance at large amplitude. We see no evidence of an instability at large amplitudes.

III. CONCLUDING REMARKS

We have illustrated here that a finite chain of appropriate spins possesses a rich spectrum of intrinsic nonlinear spin excitations. These are stable against small perturbations, so far as we can see. If an external rf field is applied, we still find these states with properties modified from the zero field case as described. As the strength of the external field is increased, these states become unstable with respect to small perturbations, but they have a lifetime that can be quite long, even for appreciable field strengths. We may view these as long-lived nonlinear resonances of the system.

There are analogies in our mind between the intrinsic nonlinear spin excitations explored here and the gap solitons described in the literature on nonlinear dielectric superlattices.[12–15] These are soliton states which exist within gaps of the dispersion curves for linear electromagnetic wave propagation down the structure. The infinitely long structure exhibits envelope solitons for such frequencies. The finite superlattice has a sequence of multisoliton states at one single frequency (with finite lifetime by virtue of radiative damping from the ends. The analogue of radiative damping is absent from our spin system).

In the case of the nonlinear dielectric structure, with nonlinearity of the Kerr form, it has proved possible to solve the Maxwell equations for a finite structure with nonlinear response characteristics.[12,14,15] As the amplitude of the input is increased, one finds a sequence of transmission resonances, associated with the excitation of one soliton, two soliton, . . . , states.

We believe that similar behavior should be exhibited by magnetic superlattices described by the effective Hamiltonian in Eq. (1), provided the exciting field is rendered spatially inhomogeneous either because its wavelength is comparable to the size of the unit cell or possibly by the presence of a finite skin depth. To explore this question, one must solve Eq. (2), now with the field amplitude h_n coupled to Maxwell's equations. This is a formidable problem, because the spin system's response is not only nonlinear, but nonlocal in space by virtue of the exchange couplings.

We regard the question of generating or detecting the nonlinear spin excitations discussed here as most fascinating. It would be of interest to see experiments on magnetic superlattices, but the metallic character of many systems studied presently renders studies with intense fields problematical unless a pulsed mode of operation could be employed.

ACKNOWLEDGMENT

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