Spin chains with a periodic array of impurities

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We investigate a spin-chain model composed of a periodic array of two kinds of spins S_1 and S_2, which allows us to study the spin chains with impurities, as well as the alternating spin chains, in a unified fashion. By using the Lieb-Shultz-Mattis theorem, we first study the model rigorously, and then by mapping it to the nonlinear σ model, we extensively investigate low-energy properties with particular emphasis on the competition between the massive and massless phases. [S0163-1829(97)51322-1]

Quantum spin chains have been providing a number of hot topics not only in condensed-matter physics but also in statistical physics, quantum field theory, etc. In particular, the competition between the massive and massless states in the spin chains has attracted much current interest. For example, doping impurities into massive systems such as the Haldane-gap system,[1] the two-leg ladder system,[2,3] and the spin-Peierls system[4] may cause the massless states, and sometimes stabilize the long-range magnetic order. For these systems it is an interesting issue to clarify how the quantum coherence, which produces spin gaps, is suppressed by impurities to result in a gapless state.[5] Also, if the impurities are magnetic and their concentration becomes high with periodic arrangement, the system naturally leads to the alternating spin chains which have also been studied intensively.[6] A common feature in these problems is how the massive and massless states compete with each other, providing a variety of interesting phenomena.

In this paper we investigate the quantum spin chain composed of the periodic array of two kinds of spin S_1 and S_2, putting particular emphasis on the formation of the massive and massless states. The model proposed here is related to the interesting topics mentioned above, and naturally interpolates the impurity models and the alternating spin-chain models. For example, if we consider the case of dilute S_1 spins in the background of the host spins with integer S_2, the model is related to the Haldane-gap systems with magnetic impurities. On the other hand, in the high-density limit of S_1 spins, it describes the alternating spin chains. We will investigate low-energy properties of the model by using the Lieb, Schultz and Mattis (LSM) theorem and nonlinear σ model techniques.

The model we will investigate consists of two kind of spins with nearest-neighbor interaction. The Hamiltonian is given by

$$H=\sum_{j=1}^{N} J_j \boldsymbol{S}_j\cdot \boldsymbol{S}_{j+1}, \tag{1}$$

with the periodic boundary condition $\boldsymbol{S}_{j+N}=\boldsymbol{S}_j$, where $N(=MN')$ is the number of sites, which is assumed to be even integral in what follows. The spin quantum number S_j on each site is $S_j=S_1$ for $j=1$ (mod M) and $S_j=S_2$ for others. The periodic array of S_1 impurities is embedded in the host spins S_2 with the period M, i.e.,

$$\underbrace{S_1\otimes S_2\otimes S_2\otimes\cdots\otimes S_2}_{M}\otimes S_1\otimes S_2\otimes\cdots\cdots\otimes S_2. \tag{2}$$

Therefore by tuning the period M, we can naturally interpolate the dilute impurity models (large M) and the alternating spin chains (M being order of unity). The antiferromagnetic coupling $J_j(>0)$ is assumed to have a bond dependence, $J_j=J(1+\gamma)$ for $j=0$ and 1 (mod M; between spins with $S=S_1$ and S_2) and $J_j=J$ for others (between the same spins with $S=S_2$).

Let us start by specifying the ground-state properties. Since spins are on a bipartite lattice, we can apply the Marshall theorem[7] to Eq. (1): The ground state is specified by the spin quantum number $S=0$ ($|S_1-S_2|N/M$) for $M=$ odd (even), which is nondegenerate except for the trivial spin degeneracy. Therefore the ground state of our model is either a spin singlet or ferrimagnetic. We are interested in the quantum effects on the spin-liquid phase, so that we will mainly concentrate on the singlet cases (odd M) which possess a variety of interesting properties. We shall show the simple but remarkable fact that the universality class of the model is solely determined by the impurity spin S_1, as long as the concentration of S_1 spins is finite.

To begin with, we apply the LSM theorem[8] in order to address the question of whether the above model can have a gapless excitation. Let T^M be the M-sites translation operator, which commutes with the Hamiltonian (1) by definition. Therefore, together with the Marshall theorem, we find that it acts on the ground-state $|\Phi_0\rangle$ as $T^M|\Phi_0\rangle=e^{i\phi}|\Phi_0\rangle$. Next we define the twist operator $U=\exp[(2\pi i/N)\Sigma_{j=1}^N jS_j^z]$, and the corresponding twisted state $|\Phi\rangle=U|\Phi_0\rangle$. The energy increment due to twist is easily calculated as $\langle\Psi|H|\Psi\rangle-\langle\Psi_0|H|\Psi_0\rangle<\text{const}/N$. In order to ensure that the twisted state $|\Phi\rangle$ is actually an excited state, we need to show the orthogonality of these states. We immediately find $T^M U T^{-M}=(-1)^{2S_{\text{eff}}M}U$ with

$$S_{\text{eff}}=[S_1+(M-1)S_2]/M. \tag{3}$$

Note that the orthogonality condition is satisfied by the minus sign of the factor $(-1)^{2S_{\text{eff}}M}$. Therefore, in the case

$2S_{\rm eff}M=$ odd integer, we can prove that the twisted state is orthogonal to the ground state $\langle\Phi_0|\Phi\rangle=0$, and hence the system has a gapless excitation, or alternatively, degenerate ground states. For example, this is the case for the spin chain with $S_1=1/2$ and $S_2=1$. In general, the existence of an excited state with $O(1/N)$ energy depends on how many half-integer spins are included in the unit composed of M sites.

Here we deal with the most interesting case, i.e., the integer S_2 spin model with S_1 spins doped, which may be related to the Haldane-gap system with magnetic impurities. According to the above theorem, it is seen in this case: it is the impurity spin S_1 that controls whether the system is massless (or massive with degenerate ground states) in the integer S_2 background. For dilute impurities with half-integer S_1, an excited state with energy of $O(1/N)$ ensured by the LSM theorem may be essentially the same as an impurity state in the Haldane-gap systems. According to extensive work on the Haldane-gap systems, doping impurities induces the free $S=1/2$ degrees of freedom[9] near the edge of the valence-bond-solid states,[10] and they couple with the doped impurity spin S_1, making a local object of the effective impurity state.[11] If we increase the concentration of S_1 spins, correlations among these local impurity states become strong, and impurity bands are naturally formed by coherent motion of effective impurity spins. Owing to the quantum effects, they again become massless (massive, see below) for half-integer (integer) S_1, which will be discussed below in terms of field-theoretic methods.

Based on the above rigorous results, we now construct a low-energy effective-field theory, which allows us to study the model with integer-S_1 as well as half-integer-S_1 cases in a unified way. Moreover, we can see which possibility for the ground state is realized for half-integer-S_1 cases, i.e., a massless state or a massive state with a degenerate ground state. For this purpose, we shall map the system (1) to the nonlinear σ model[12,13] by the use of the SU(2) coherent state path integrals.[14] In what follows, we again concentrate on the systems with a singlet ground state, i.e., for the case $M=$ odd. Note that the following analysis can be applied to the system with *finite concentration* of impurities in the thermodynamic limit.

The coherent state in the spin-S representation is here defined by $|\zeta\rangle=(1+|\zeta|^2)^{-S}\exp(\zeta S^-)|S,m=S\rangle$. Parametrizing $\zeta=\tan(\theta/2)\,e^{i\phi}$, we have $\langle\zeta|\mathbf{S}|\zeta\rangle=S\mathbf{n}$, where $\boldsymbol{n}=(\sin\theta\cos\phi,\sin\theta\sin\phi,\cos\theta)$. By using the overcompleteness relation $\int d\mu(\zeta)|\zeta\rangle\langle\zeta|=1$ with the invariant measure $d\mu(\zeta)=(2S+1)\pi/(1+|\zeta|^2)^2d^2\zeta$, we can derive the path-integral representation of the partition function. Namely, by staggering the spin configuration as $\boldsymbol{S}_j=S_j(-)^{j+1}\boldsymbol{n}(j)$, the partition function $Z=\mathrm{tr}\exp(-\beta H)$ of the system (1) can be represented by $Z=\int\Pi_{j=1}^N\mathcal{D}\mu[\boldsymbol{n}(j)]\exp(-S)$, where the action is explicitly given by

$$S=i\sum_{j}^{N}(-)^jS_j\omega[\boldsymbol{n}(j)]-\sum_{j=1}^{N}J_jS_jS_{j+1}\int_0^\beta d\tau\boldsymbol{n}(j)\cdot\boldsymbol{n}(j+1). \tag{4}$$

Here $\omega[\boldsymbol{n}(j)]$ is the Berry phase acquired by the jth spin,

$$\omega[\boldsymbol{n}(j)]=\int_0^1du\int_0^\beta d\tau\boldsymbol{n}(j)\cdot[\partial_\tau\boldsymbol{n}(j)\times\partial_u\boldsymbol{n}(j)]. \tag{5}$$

In what follows, we assume that the behavior of the field $\boldsymbol{n}(j)$ is similar to that for the uniform spin chain in the continuum limit. Therefore, we can divide $\boldsymbol{n}(j)$ into the slowly varying part and fluctuation around it as usual, $\boldsymbol{n}(j)=\boldsymbol{m}(j)+a(-)^{j+1}\boldsymbol{l}(j)$. This assumption should be confirmed to be valid by comparison with the results of the LSM theorem.

Let us first calculate the Berry phase term in the continuum limit,

$$\begin{aligned}S_B&=i\sum_{k=1}^{M}\sum_{j=1}^{N/(2M)}[S_1\delta_{k,1}-S_2(-)^k(1-\delta_{k,1})]\\&\quad\times\{\omega[\boldsymbol{n}(2Mj-M+k)]-\omega[\boldsymbol{n}(2Mj-2M+k)]\}\\&=i\frac{S_1}{2}\int\int d^2x\boldsymbol{m}\cdot(\partial_1\boldsymbol{m}\times\partial_2\boldsymbol{m})\\&\quad+iS_{\rm eff}\int\int d^2x\boldsymbol{l}\cdot(\boldsymbol{m}\times\partial_2\boldsymbol{m})\end{aligned} \tag{6}$$

with $S_{\rm eff}$ defined in Eq. (3), where $x_1=x$ and $x_2=\tau$. To derive this formula, we have taken the continuum limit with respect to Ma lattice spacing: $\boldsymbol{n}(2Mj-M+k)-\boldsymbol{n}(2Mj-2M+k)\sim Ma\partial_1\boldsymbol{m}(2Mj-M+k)+(-)^k2a\boldsymbol{l}(2Mj-M+k)$, where a is the lattice constant. This procedure implies that we are now concerned with the most important low-energy mode, although there are other massive excitation modes because the period of the lattice is M in our model. The validity of this procedure will be discussed by comparing the results with those of the LSM theorem. It should also be noted that the continuum limit here corresponds to the limit $N/(2M)\to\infty$. Namely, we assume in this approach the finite concentration $1/M$ of the S_1 spins. Next, the interaction term is rewritten in the continuum limit,

$$\begin{aligned}S_I&=-J\sum_{k=1}^{M}\sum_{j=1}^{N/M}[(1+\gamma)S_1S_2(\delta_{k,1}+\delta_{k,M})+S_2^2(1-\delta_{k,1})\\&\quad\times(1-\delta_{k,M})]\int d\tau\mathbf{n}(Mj-M+k)\cdot\boldsymbol{n}(Mj-M+k+1)\\&\sim\frac{Ja}{2}\beta\int\int d^2x[(\partial_1\boldsymbol{m})^2+4\boldsymbol{l}^2]+\mathrm{const}\end{aligned} \tag{7}$$

with $\beta=S_2^2+2[(1+\gamma)S_1-S_2]S_2/M$.[15] In this way, we end up with the action composed of the fields $\boldsymbol{m}(x)$ and $\boldsymbol{l}(x)$. The term of $\boldsymbol{l}(x)$ appears in a quadratic form, and can be easily integrated out, which consequently results in the following Lagrangian density

$$\mathcal{L}=\frac{1}{2g}\left[v(\partial_1\boldsymbol{m})^2+\frac{1}{v}(\partial_2\boldsymbol{m})^2\right]+\frac{\theta}{8\pi}\epsilon_{\mu\nu}\boldsymbol{m}\cdot(\partial_\mu\boldsymbol{m}\times\partial_\nu\boldsymbol{m}), \tag{8}$$

with

$$\theta=2\pi i S_1 \quad (=2\pi i S_{\rm eff} M),$$

$$g=\frac{2}{S_2+(S_1-S_2)/M} \quad \left(=\frac{2}{S_{\rm eff}}\right),$$

$$v=2Ja\,\frac{S_2^2+2[(1+\gamma)S_1-S_2]S_2/M}{S_2+(S_1-S_2)/M}, \tag{9}$$

where $S_{\rm eff}$ is defined in Eq. (3).

We recall here that the topological term with θ can completely specify whether the system is massive or massless. By observing that only S_1 appears in θ, we again arrive at the conclusion that *the universality class of the system is solely determined by the impurity spin* S_1, which completely fits in with the results of the extended LSM theorem obtained above: for half-integer S_1, the system is massless even if $S_2=$ integer because $\theta=\pi i$ (mod $2\pi i$). Furthermore, for integer S_1 we can say beyond the LSM theorem that the system should be massive because $\theta=0$ (mod $2\pi i$). By using the above formulas, we can discuss the interesting results deduced by the LSM theorem in more detail. The following statements are valid for the system with a finite concentration of impurities. (a) Haldane-gap systems ($S_2=$ integer) become massless (are still massive) for half-integer (integer) S_1 impurities. (b) Massless spin chains ($S_2=$ half-integer) become massive (are still massless) for integer (half-integer) S_1 impurities. Especially in the latter case, we would like to recall the analysis by Eggert and Affleck,[16] from which we can naively expect the following scenario: Integer S_1 impurities are screened by the two neighboring half-integer spins S_1, forming local integer S_2-2S_1 objects. These effective spins may couple with each other in a coherent way and produce a gap in the way suggested by Haldane.

We should mention here that our mapping to the σ model may be justified for a high or intermediate concentration of impurities, but not for a dilute limit. However, a qualitative feature of whether the system is massive or not is determined solely by the topological term, and is expected to be correctly specified via the present analysis so far as the impurity concentration is finite in the thermodynamic limit.

So far we have been mainly concerned with the impurity effects on spin chains. Here we discuss the opposite limiting case (high density of S_1 spins), i.e., the alternating spin-chain system in more detail. For example, when $M=2$ with a half-integer S_1 and an integer S_2, our model (1) neatly describes the alternating spin-chain system which has a ferrimagnetic ground state,[17] as being consistent with experiments found so far.[6] In this connection, modified alternating spin chains with a singlet ground state (quantum liquid phase) have been actively investigated, for which quantum fluctuations should play a vital role.[18,19] This problem may provide a new, interesting paradigm of spin chains bridging the massive and massless Heisenberg chains. As a typical example,[19] we here consider a slight extension of the Hamiltonian (1) with $S_j=S_1$ for $j=1$ and 2 (mod 4) and $S_j=S_2$ for $j=3$ and 4 (mod 4), namely,

$$S_1\otimes S_1\otimes S_2\otimes S_2\otimes S_1\otimes S_1\otimes\cdots\otimes S_2\otimes S_2. \tag{10}$$

Antiferromagnetic couplings are $J_j=J(1-\gamma_1)$ for $j=1$ (mod 4; between S_1), $J_j=J(1-\gamma_2)$ for $j=3$ (mod 4; between S_2) and $J_j=J[1+(\gamma_1+\gamma_2)/2]$ for $j=$ others (between S_1 and S_2). According to the Marshall theorem, it is easily found that the ground state of this model is singlet. Though the system (10) is invariant under four-sites translation T^4, the LSM theorem cannot apply for any values of S_1 and S_2, i.e., the twisted state is not orthogonal to the ground state. This suggests that the system may be massive in general. However, we should also note that since the system has spin alternation as well as bond alternation, there could exist nontrivial massless phases between the massive phases, as is the case for uniform spin systems.[20] We will clarify these points using nonlinear σ model techniques.

Keeping the above observations in mind, we derive the effective theory. We have the same Lagrangian (8), but with

$$\theta=2\pi i S_{\rm eff}(1+\gamma_{\rm eff}),$$

$$g=\frac{2}{S_{\rm eff}\sqrt{1-\gamma_{\rm eff}^2}},$$

$$v=2Ja(S_{\rm eff}-\Delta S_\gamma)\sqrt{1-\gamma_{\rm eff}^2}, \tag{11}$$

where

$$S_{\rm eff}=(S_1+S_2)/2,$$

$$\Delta S_\gamma=(S_1-S_2)(\gamma_1 S_1-\gamma_2 S_2)/(4S_{\rm eff}),$$

$$\gamma_{\rm eff}=\frac{(S_1+S_2)(\gamma_1 S_1+\gamma_2 S_2)-(S_1-S_2)^2}{(S_1+S_2)^2-(S_1-S_2)(\gamma_1 S_1-\gamma_2 S_2)}. \tag{12}$$

We wish to study nontrivial cases, taking $S_1=1/2$ and $S_2=1$ as an example, and then generalize the discussions to arbitrary spin cases. To begin with, let us set $\gamma_1=\gamma_2=0$. We find from Eqs. (11) and (12) that the topological term θ is different from πi (mod $2\pi i$), and therefore the system is massive. Even in this case, $\gamma_{\rm eff}\neq 0$ since it includes the effects of the spin alternation. Let us next introduce the bond-alternation term γ, and observe what happens if the parameter γ changes from -1 to 1 for the case $\gamma_1=\gamma_2\equiv\gamma$. At $\gamma=-1$, the model (10) becomes a set of isolated dimers, as seen from (10). This system has massive excitations, which is indeed consistent with $\theta=0$ and $g\to\infty$ in Eq. (11). If we increase γ up to 1, the effective $\gamma_{\rm eff}$ changes from $\gamma_{\rm eff}=-1$ to 1. The corresponding value of θ changes from 0 to $\theta=3\pi i$. So it is predicted that we encounter a massless fixed point once at $\theta=\pi i$ during this process. For the case $\gamma\to 1$, we need more careful treatment. As naively expected, in this case, S_1 and S_2 spins strongly couple with each other antiferromagnetically and the model (10) should behave like the uniform $S=1/2$ spin chain with massless excitations (indeed we have $\theta\to 3\pi i$). We should note, however, that exactly at $\gamma=1$ the system is separated into isolated pairs of spins, which may cause a singular behavior in the coupling constant, $g\to\infty$. Therefore, for $\gamma\to 1$ the model exhibits a behavior quite similar to that for a half-integral spin chain although it is still massive.

In general, for the case with half-odd integer S_1 and integer S_2, we can predict that there appear massless phases $2S_{\rm eff}-1/2$ times as the bond alternation parameter changes from $\gamma=-1$ to 1. It is also shown that for the cases where

S_1 and S_2 are the same type of spins, i.e., half-odd integers or integers, there appear $2S_{\rm eff}$ times massless fixed points. We wish to note that in the uniform case $S_1=S_2=S_{\rm eff}$ our formulas reduce to those for the ordinary spin chain with bond alternation, which has $2S_{\rm eff}$ critical points for $-1<\gamma<1$.[20] In this way, our approach based on nonlinear σ model techniques are quite powerful to systematically study the low-energy properties of alternating spin-chain systems.

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[1] M. Hagiwara, K. Katsumata, I. Affleck, B. I. Halperin, and J. P. Renard, Phys. Rev. Lett. **65**, 3181 (1990); S. H. Glarum, S. Geschwind, K. M. Lee, M. L. Laplan, and J. Michel, *ibid.* **67**, 1614 (1991).

[2] T. M. Rice, S. Gopalan, and M. Sigrist, Europhys. Lett. **23**, 445 (1993); E. Dagotto and T. M. Rice, Science **271**, 618 (1996).

[3] M. Nohara, H. Takagi, M. Azuma, Y. Fujishiro, and M. Takano (unpublished); M. Azuma, Y. Fujishiro, M. Takano, T. Ishida, K. Okuda, M. Nohara, and H. Takagi (unpublished).

[4] M. Hase, I. Terasaki, and K. Uchinokura, Phys. Rev. Lett. **70**, 3651 (1993); L. P. Regnault, J. P. Renard, G. Dhalenne, and A. Revcolevschi, Europhys. Lett. **32**, 579 (1995).

[5] H. Fukuyama, T. Tanimoto, and M. Saito, J. Phys. Soc. Jpn. **65**, 1183 (1996); H. Fukuyama, N. Nagaosa, M. Saito, and T. Tanimoto, *ibid.* **65**, 2377 (1996); M. Sigrist and A. Furusaki, *ibid.* **65**, 2385 (1996); G. Martins, E. Dagotto, and J. Riera, Phys. Rev. B **54**, 16 032 (1996);H.-J. Mikeska, U. Neugebauer, and U. Schollwöck, *ibid.* **55**, 2955 (1997); N. Nagaosa, A. Furusaki, M. Sigrist, and H. Fukuyama, *ibid.* **51**, 15 588 (1995); Y. Motome, N. Katoh, N. Furukawa, and M. Imada, J. Phys. Soc. Jpn. **65**, 1949 (1996); Y. Iino and M. Imada (unpublished); T. Fukui, M. Sigrist, and N. Kawakami, Phys. Rev. B (to be published).

[6] G. T. Yee, J. M. Manriquez, D. A. Dixon, R. S. McLean, D. M. Groski, R. B. Flippen, K. S. Narayan, A. J. Epstein, and J. S. Miller, Adv. Mater. **3**, 309 (1991); Inorg. Chem. **22**, 2624 (1983); **26**, 138 (1987).

[7] See, for example, A. Auerbach, *Interacting Electrons and Quantum Magnetism* (Springer-Verlag, Berlin, 1994).

[8] E. H. Lieb, T. Schultz, and D. J. Mattis, Ann. Phys. (N.Y.) **16**, 407 (1961).

[9] T. Kennedy, J. Phys., Condens. Matter. **2**, 5737 (1990).

[10] I. Affleck, T. Kennedy, E. H. Lieb, and H. Tasaki, Phys. Rev. Lett. **59**, 799 (1987).

[11] E. S. Sørensen and I. Affleck, Phys. Rev. B **51**, 16 115 (1995); Z.-Y. Lu, Z.-B. Su, and L. Yu, Phys. Rev. Lett. **74**, 4297 (1995); K. Penc and H. Shiba, Phys. Rev. B **52**, R715 (1995).

[12] F. D. M. Haldane, Phys. Lett. **93A**, 464 (1983); Phys. Rev. Lett. **50**, 1153 (1983).

[13] For reviews, see I. Affleck, in *Field Theory Methods and Quantum Critical Phenomena*, Les Houches Lecture Series, edited by E. Brézin and J. Zinn-Justin (North-Holland, Amsterdam, 1990); E. Fradkin, *Field Theories of Condensed Matter Physics* (Addison-Wesley, Reading, MA, 1994); A. M. Tsvelik, *Quantum Field Theory in Condensed Matter Physics* (Cambridge University Press, Cambridge, England, 1995).

[14] J. R. Klauder, Phys. Rev. D **19**, 2349 (1979); H. Kuratsuji and T. Suzuki, J. Math. Phys. (N.Y.) **21**, 472 (1980).

[15] This β is valid only for $M\geqslant 3$. If we include $M=1$, we should replace it by $\beta=S_1^2\delta_{M,1}+\{S_2^2+2[(1+\gamma)S_1-S_2]S_2/M\}(1-\delta_{M,1})$.

[16] S. Eggert and I. Affleck, Phys. Rev. B **46**, 10 866 (1992).

[17] S. K. Pati, S. Ramasesha, and D. Sen, Phys. Rev. B **55**, 8894 (1997); A. K. Kolezhuk, H.-J. Mikeska, and S. Yamamoto, *ibid.* **55**, 3336 (1997); H. Niggemann, G. Uimin, and J. Zittartz (unpublished).

[18] H. J. de Vega and F. Woynarovich, J. Phys. A **25**, 449 (1992); M. Fujii, S. Fujimoto, and N. Kawakami, J. Phys. Soc. Jpn. **65**, 2381 (1996).

[19] T. Tonegawa, M. Kaburagi, and S. Miyashita (unpublished).

[20] I. Affleck and F. D. M. Haldane, Phys. Rev. B **36**, 5291 (1987).