Nonlinear σ Model for Inhomogeneous Spin Chains

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We derive a nonlinear σ model (NLSM) that represents antiferromagnetic Heisenberg spin chains with inhomogeneous spin magnitudes and inhomogeneous nearest-neighbor exchange constants arrayed in finite periods. The only restriction is that the average spin magnitude on a sublattice is the same as that on another sublattice. The NLSM yields the gapless condition in terms of the spin magnitudes and exchange constants. We apply this condition to several cases, including systems with impurities. The result shows that the spin gap persists, despite impurities, in many cases. [S0031-9007(99)09421-1]

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In 1983 Haldane predicted that a homogeneous antiferromagnetic Heisenberg spin chain is gapful if the spin magnitude is an integer, and is gapless if it is a half-odd integer [1]. Since his theory is based on mapping of a spin model to the nonlinear σ model (NLSM), the importance of the NLSM in condensed matter physics is recognized; various interesting aspects of the NLSM method are seen in Refs. [2–4]. Affleck [5] reformulated the NLSM method in an operator formalism. He divided a spin chain into spin pairs and transformed the spin operators for each spin pair into operators representing an antiferromagnetic motion and a small fluctuation. The operator formalism is applicable even to a spin chain with bond alternation, which is the simplest model with inhomogeneity for the exchange constant. It shows that, e.g., a spin chain with magnitude $1/2$ has a gapless spin excitation only at the phase boundary point between dimerized phases.

Generally, how does the inhomogeneity affect the spin gap in an arbitrary spin chain? To answer this question we need to obtain the gapless condition for various inhomogeneous spin chains. There are two kinds of inhomogeneity in a spin chain: one is for the exchange constant and the other is for the spin magnitude. It is difficult to extend the operator formalism of Affleck to various inhomogeneous cases, especially to cases where the spin magnitude is inhomogeneous; it would need a very complicated transformation among operators. Fukui and Kawakami tried to extend an NLSM method to some special inhomogeneous chains in a path integral formalism [6,7]. They rather intuitively introduced favorable variables to form an NLSM and did not care to preserve the degrees of freedom. Hence, although some of their results seem to be plausible, we cannot determine whether they are really correct or not by inspecting their theory itself. The correct and general method for systems with inhomogeneous spin magnitude has not been established.

In this Letter, we unambiguously derive a general NLSM representing a wide class of inhomogeneous antiferromagnetic spin chains. For this purpose we divide the system into blocks of the same size, and transform the variables of integration for each block with preserving the original degrees of freedom in a path integral formula. We regard a spin pair in Affleck's method [5] as an especially simple block. From the NLSM we obtain the gapless condition in a simple closed form. We apply this result to several cases including systems with impurities.

We consider the spin chain represented by the Hamiltonian

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

where $\mathbf{S}_j$ is the spin at site j and $J_j(>0)$ is the exchange constant between $\mathbf{S}_j$ and $\mathbf{S}_{j+1}$. The number of lattice sites is N, the lattice spacing is a, and the system size is $L = aN$. The quantum number for the magnitude of $\mathbf{S}_j$ is denoted by s_j. The system is periodic with period $2b$ (b: a positive integer):

$$J_{j+2b} = J_j, \qquad s_{j+2b} = s_j\,. \tag{2}$$

We divide the spin chain into blocks each of which contains $2b$ spin sites. The block size $2ba$ is an arbitrary (even) positive integer times the size of the unit cell if a unit cell contains an even (odd) number of spins. In a block $\{J_j\}$ and $\{s_j\}$ are arbitrary except for the following restriction, which excludes systems with ferrimagnetic ground states in the classical limit:

$$\sum_{j=1}^{b} s_{2j} = \sum_{j=1}^{b} s_{2j-1}\,. \tag{3}$$

The partition function Z is written in a path integral formula. When $(-1)^j s_j \mathbf{n}_j$ is the expectation value of $\mathbf{S}_j$ for a coherent state, it is

$$Z = \int D[\mathbf{n}_j] \prod_j \delta(\mathbf{n}_j^2 - 1) e^{-S}. \tag{4}$$

The action S at temperature $1/\beta$ is given by

$$S = i \sum_{j=1}^{N} (-1)^j s_j w[\mathbf{n}_j] + \frac{1}{2} \int_0^\beta d\tau \sum_{j=1}^{N} \tilde{J}_j (\delta \mathbf{n}_j)^2 \tag{5}$$

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with $\delta \mathbf{n}_j = \mathbf{n}_{j+1} - \mathbf{n}_j$ and

$$\tilde{J}_j = J_j s_j s_{j+1}. \tag{6}$$

The first term in Eq. (5) comes from the Berry phase and $w[\mathbf{n}_j]$ is the solid angle which the unit vector $\mathbf{n}_j$ forms in the period β.

We transform spin variables $\{\mathbf{n}_j\}$ into gradually changing variables $\{\mathbf{m}(p)\}$ and small fluctuations $\{\mathbf{L}_q(p)\}$, where p labels a block in the lattice ($p = 1, 2, \ldots, N/2b$) and q labels a site in the block ($q = 1, 2, \ldots, 2b$). The spin variable at the qth site in the pth block is written as

$$\mathbf{n}_{2bp+q} = (1 - z_q)\mathbf{m}(p) + z_q \mathbf{m}(p - \gamma_q) + a\mathbf{L}_q(p) \tag{7}$$

with $z_q = |b - q|/2b$ and $\gamma_q = \mathrm{sgn}(b - q + \frac{1}{2})$. The original constraints $\{\mathbf{n}_j^2 = 1\}$ are changed to $\{\mathbf{m}^2(p) = 1\}$ and $\{\mathbf{m}(p) \cdot \mathbf{L}_q(p) = 0\}$. Here we notice that the number of variables increases in this transformation. To solve the problem we can add an additional constraint for each block which is arbitrary as long as it consists with the other constraints; e.g., $\sum_{q=1}^{2b} \mathbf{L}_q(p) = 0$ or $\mathbf{L}_{2b}(p) = 0$ is a possible one. Hence $2b$ vector variables $\{\mathbf{n}_{2bp+q} \,|\, q = 1, 2, \ldots, 2b\}$ per block are transformed to $2b$ vector variables $\{\mathbf{m}(p), \mathbf{L}_q(p) \,|\, q = 1, 2, \ldots, 2b - 1\}$ per block; $\mathbf{L}_{2b}(p)$ is written by the other variables through the additional constraint. Also $2b$ original constraints $\{\mathbf{n}_q^2 = 1 \,|\, q = 1, 2, \ldots, 2b\}$ per block are replaced by $2b$ constraints $\{\mathbf{m}^2(p) = 1, \mathbf{m}(p) \cdot \mathbf{L}_q(p) = 0 \,|\, q = 1, 2, \ldots, 2b - 1\}$ per block. Thus we obtained a new set of variables without changing the original degrees of freedom in the path integral formula (4). In other words, we transformed only the variables of integration. As for the additional constraint, the choice does not affect physical quantities at all. This is because the variables $\{\mathbf{L}_q(p) \,|\, q = 1, 2, \ldots, 2b\}$ appear only as $\{\mathbf{L}_q(p) - \mathbf{L}_{q+1}(p) \,|\, q = 1, 2, \ldots, 2b - 1\}$ in the partition function, as will be seen.

To take a continuum limit we identify the center of the pth block, $(2bp + b)a$, as coordinate x. Then the difference between adjacent spin variables is replaced as

$$\delta \mathbf{n}_{2bp+q} \to a[\partial_x \mathbf{m}(x) - \mathbf{R}_q(x)], \tag{8}$$

$$\mathbf{R}_q(x) = \mathbf{L}_q(x) - \mathbf{L}_{q+1}(x). \tag{9}$$

Equation (9) for $q = 2b$ reads as $\mathbf{R}_{2b}(x) = \mathbf{L}_{2b}(x) - \mathbf{L}_1(x)$. In the Berry phase term of the action (5), the following relation stands due to the restriction (3):

$$\sum_{q=1}^{2b} (-1)^q s_q w[\mathbf{n}_{2bp+q}] = \sum_{q=1}^{2b} \tilde{s}_q \delta w[\mathbf{n}_{2bp+q}], \tag{10}$$

where $\delta w[\mathbf{n}_{2bp+q}] = w[\mathbf{n}_{2bp+q+1}] - w[\mathbf{n}_{2bp+q}]$ and

$$\tilde{s}_q = \sum_{k=1}^{q} (-1)^{k+1} s_k. \tag{11}$$

The continuum limit of the Berry phase term is taken after $\delta w[\mathbf{n}_{2bp+q}]$ is transformed to the τ integral of $\delta \mathbf{n}_{2bp+q} \cdot (\mathbf{n}_{2bp+q} \times \partial_\tau \mathbf{n}_{2bp+q})$ as in the usual way [3,4]. Thus the action (5) becomes $S_c = S_1 + S_2$ with

$$S_1 = \int_0^\beta d\tau \int_0^L dx \left\{ -i \frac{s'}{2} (\partial_x \mathbf{m}) \cdot (\mathbf{m} \times \partial_\tau \mathbf{m}) + \frac{a}{2} \bar{J} (\partial_x \mathbf{m})^2 \right\}, \tag{12}$$

$$S_2 = \int_0^\beta d\tau \int_0^L dx \, \frac{1}{2b} \sum_{q=1}^{2b} \left(\frac{a}{2} \tilde{J}_q \mathbf{R}_q^2 + i \mathbf{f}_q \cdot \mathbf{R}_q \right), \tag{13}$$

where $s' = \sum_{q=1}^{2b} \tilde{s}_q / b$, $\bar{J} = \sum_{q=1}^{2b} \tilde{J}_q / 2b$, and $\mathbf{f}_q = ia\tilde{J}_q (\partial_x \mathbf{m}) + \tilde{s}_q (\mathbf{m} \times \partial_\tau \mathbf{m})$.

The variables $\{\mathbf{L}_q\}$ appear only as $\{\mathbf{R}_q\}$ in the action S_c. The variables $\{\mathbf{R}_q\}$ are not independent and equation $\sum_{q=1}^{2b} \mathbf{R}_q = 0$ stands due to the definition (9); e.g., we can delete $\mathbf{R}_{2b}$ by the equation. We treat the equation as a new constraint. The constraints $\{\mathbf{m} \cdot \mathbf{L}_q = 0\}$ are rewritten as $\{\mathbf{m} \cdot \mathbf{R}_q = 0\}$. Instead of deleting some variables by constraints, we insert the corresponding δ functions into the path integral formula (4) and treat all the variables independently. We use the integral representations of $\delta(\sum_{q=1}^{2b} \mathbf{R}_q)$ and $\delta(\mathbf{m} \cdot \mathbf{R}_q)$ with integration variables $\mathbf{u}$ and α_q. Then the following term appears in addition to the action S_c:

$$S_3 = \int_0^\beta d\tau \int_0^L dx \, \frac{i}{2b} \sum_{q=1}^{2b} \mathbf{R}_q \cdot (-\mathbf{u} + \alpha_q \mathbf{m}). \tag{14}$$

Carrying out integrations in the partition function first with respect to $\{\mathbf{R}_q\}$ and then to $\mathbf{u}$, $S_2 + S_3$ reduces to

$$S_2' = \int_0^\beta d\tau \int_0^L dx \, \frac{1}{4ba} \sum_{q=1}^{2b} \frac{1}{\tilde{J}_q} (\mathbf{F}_q^2 - \bar{\mathbf{F}}^2), \tag{15}$$

where $\mathbf{F}_q = \mathbf{f}_q + \alpha_q \mathbf{m}$ and

$$\bar{\mathbf{F}} = \sum_{q=1}^{2b} (\tilde{J}_q)^{-1} \mathbf{F}_q \Big/ \sum_{q=1}^{2b} (\tilde{J}_q)^{-1}. \tag{16}$$

Expanding the integrand in Eq. (15), we find that $\{\alpha_q\}$ appear only in a bilinear form and are integrated out.

Collecting S_1 and the remnant of S_2' after the integration with respect to $\{\alpha_q\}$, we have the final effective action:

$$S_{\mathrm{eff}} = \int_0^\beta d\tau \int_0^L dx \left\{ -i \frac{J^{(0)}}{J^{(1)}} \mathbf{m} \cdot (\partial_\tau \mathbf{m} \times \partial_x \mathbf{m}) + \frac{1}{2aJ^{(1)}} \left(\frac{J^{(1)}}{J^{(2)}} - \frac{J^{(0)}}{J^{(1)}} \right) (\partial_\tau \mathbf{m})^2 + \frac{a}{2} J^{(0)} (\partial_x \mathbf{m})^2 \right\}, \tag{17}$$

where $\{J^{(n)}\}$ are defined as

$$\frac{1}{J^{(n)}} = \frac{1}{2b} \sum_{q=1}^{2b} \frac{(\tilde{s}_q)^n}{\tilde{J}_q} \qquad (n = 0, 1, 2) \tag{18}$$

with Eqs. (6) and (11). Thus we have obtained the action of the NLSM describing the Hamiltonian (1) in the

continuum limit. The real-space cutoff is the length of a block of the minimum size. The topological angle θ is given by setting $J^{(0)}/J^{(1)}$ in the first term as $\theta/4\pi$. The velocity v and the coupling constant g are given by equating the coefficients of $(\partial_\tau \mathbf{m})^2$ and of $(\partial_x \mathbf{m})^2$ as $1/2gv$ and $v/2g$, respectively. Note that the coefficient of $(\partial_\tau \mathbf{m})^2$ is always positive.

The action (17) is independent of the way to divide the system into blocks. First we displace each block by one site. Then the spin magnitudes in a new block are ordered as $(s_2, s_3, \ldots, s_{2b}, s_1)$ instead of $(s_1, s_2, s_3, \ldots, s_{2b})$. Denoting quantities related to the new blocks by letters with prime, we have relations $\tilde{s}'_q = s_1 - \tilde{s}_{q+1}$ for $1 \le q \le 2b - 1$ and $\tilde{s}'_{2b} = 0$. Using these relations we obtain the following transformation: $J'^{(0)} = J^{(0)}$,

$$\frac{1}{J'^{(1)}} = \frac{s_1}{J^{(0)}} - \frac{1}{J^{(1)}}, \qquad \frac{1}{J'^{(2)}} = \frac{s_1^2}{J^{(0)}} - \frac{2s_1}{J^{(1)}} + \frac{1}{J^{(2)}}. \tag{19}$$

This transformation does not change the coefficient of $(\partial_\tau \mathbf{m})^2$ in Eq. (17). The topological angle (divided by 2π) changes as

$$\frac{2J'^{(0)}}{J'^{(1)}} = 2s_1 - \frac{2J^{(0)}}{J^{(1)}}. \tag{20}$$

Here the first term, $2s_1$, is an integer and does not affect physics. The negative sign of the second term is also irrelevant. Thus the action (17) is invariant under the block displacement.

Second we inspect the effect for the action when we use the block of size $2rba$ (r: a positive integer) instead of $2ba$. The order of the spin magnitudes in a new block is then r times repetition of $(s_1, s_2, \ldots, s_{2b})$. Because of the restriction (3) we have relation $\tilde{s}'_{2jb+q} = \tilde{s}_q$ for $q = 1, 2, \ldots, 2b$ and $j = 1, 2, \ldots, r$. Using this relation we see that $J'^{(n)} = J^{(n)}$ for $n = 0, 1, 2$ and the action (17) is invariant under the block enlargement.

The NLSM has a gapless excitation when $\theta/2\pi$ is a half-odd integer. This condition is written as

$$\frac{2J^{(0)}}{J^{(1)}} = \frac{2l - 1}{2}, \tag{21}$$

where l is an arbitrary integer. In what follows we examine this condition for several cases.

We first apply the general formula (17) to the homogeneous case: $s_1 = s_2 = \cdots = s_N \equiv s$ and $J_1 = J_2 = \cdots = J_N \equiv J$. In this case Eq. (18) gives $J^{(0)} = Js^2$, $J^{(1)} = 2Js$, and $J^{(2)} = 2J$. The coefficients in Eq. (17) are simple as $\theta/4\pi = s/2, 1/2gv = 1/8aJ$, and $v/2g = aJs^2/2$. Hence Eq. (17) in this case is equivalent to the NLSM which Haldane originally considered [1].

For $b = 1$, a block contains only two spins and the restriction (3) reads as $s_1 = s_2$. Although the correct NLSM in this case has already been obtained [2,5], we restate some results based on the general formula (17). $\{J^{(n)}\}$ are calculated as $J^{(0)} = 2s^2 J_1 J_2/(J_1 + J_2)$, $J^{(1)} = 2sJ_1$, and $J^{(2)} = 2J_1$. The gapless condition (21) is then $sJ_2/(J_1 + J_2) = (2l - 1)/4$. This equation gives the single gapless point $J_2/J_1 = 1 (l = 1)$ for $s = 1/2$ and $J_2/J_1 = 1/3 (l = 1)$ for $s = 1$. Numerical calculations for $s = 1$ show that the gapless point is at $J_2/J_1 = 0.6$ [8,9] and an experiment for $[\{\mathrm{Ni(333\text{-}tet)}\,(\mu\text{-}\mathrm{N_3})\}_n](\mathrm{ClO_4})_n$ agrees with this value [10]. Hence the method of the NLSM does not always give quantitatively correct results. However the NLSM is expected to represent the essence of quantum spin systems.

For $b = 2$, the restriction (3) is now $s_1 + s_3 = s_2 + s_4$. In the case of $s_1 = s_2$, $s_3 = s_4$, $J_1 = J_3$, and $J_2 = J_4$, we have $2J^{(0)}/J^{(1)} = 2s_1 s_3 (s_1 + s_3)/(s_1^2 + s_3^2 + 2s_1 s_3 J_1/J_2)$. Following the condition (21), a gapless excitation appears at $J_2/J_1 = 4/7$ for $s_1 = 1/2$ and $s_3 = 1$, and at $J_2/J_1 = 3/7$ for $s_1 = 1/2$ and $s_3 = 3/2$. For $s_1 = 1$ and $s_3 = 2$, two gapless points are $J_2/J_1 = 4/19$ and $4/3$. Fukui and Kawakami [7] have obtained the same results [11]. Tonegawa *et al.* performed numerical calculations for $s_1 = 1/2$ and $s_3 = 1$ and obtained $J_2/J_1 = 0.77 \pm 0.01$ for the gapless point [12]. In the case of $s_1 = s_2$, $s_3 = s_4$, $J_1 = J_2$, and $J_3 = J_4$, we have $2J^{(0)}/J^{(1)} = 2s_1 s_3/(s_1 + s_3)$. Hence the gapless excitation appears if and only if $s_1 = s_3$ and the value is a half-odd integer irrespective of the values of J_1 and J_3. In the case of $s_1 = s_2 = s_3 = s_4 \equiv s$, $J_1 = 1 - \delta$, $J_3 = 1 + \delta$, and $J_2 = J_4 \equiv J$, we have $2J^{(0)}/J^{(1)} = 2sJ/(J + 1 - \delta^2)$. Chen and Hida performed numerical calculation for $s = 1/2$ and obtained a phase boundary [13]. The positive-J part of their boundary is close to $\delta = (1 - J)^{1/2}$ determined by Eq. (21) with $l = 1$.

Using the block of $b = 3$, we can deal with various systems. Here we examine the case that a unit cell contains three sites. In a block of the minimum size, the spin magnitudes and the exchange constants are ordered as $(s_1, s_2, s_3, s_1, s_2, s_3)$ and $(J_1, J_2, J_3, J_1, J_2, J_3)$. The restriction (3) is satisfied. In this case we have $2J^{(0)}/J^{(1)} = s_1 - s_2 + s_3$ irrespective of the values of J_1, J_2, and J_3. The condition (21) says that systems with one or three half-odd-integer spins in a unit cell are gapless [7,14].

We examine a bond impurity system in the general formula (17). In this system all the spin magnitudes are the same and denoted by s; $\{\tilde{s}_q\}$ are the same as those for the homogeneous case. In contrast there are two kinds of exchange constants, J_0 and J. We assume that $2h$ impurity bonds with J_0 randomly distribute among host bonds with J in a block of size $2b$. Then $(J^{(0)})^{-1}$ is calculated as $[1 + \rho_0(J/J_0 - 1)]/Js^2$ with impurity density $\rho_0 = h/b$; the randomness does not affect this quantity. For $(J^{(1)})^{-1}$ we take an ensemble average since the contribution of an impurity bond changes whether the impurity site q is even or odd. We assume that all the possible distributions occur in the equal probability and that just half of the impurity bonds are on odd sites in the average. Hence we have $(J^{(1)})^{-1} = [1 + \rho_0(J/J_0 - 1)]/2Js$. The topological angle is then given by

$\theta/2\pi = 2J^{(0)}/J^{(1)} = s$. Since θ is independent of b, the block size $2ba$ is arbitrarily large [15]. The bond impurities do not change the gapless condition of the homogeneous case [16]. Kawae *et al.* [17] argued that the $s = 1$ Haldane system $(CH_3)_4NNi(NO)_3$ (TMNIN) is described by the bond impurity model when nonmagnetic impurities Zn^{2+} are doped. They observed spin gaps at some impurity densities.

The last example is a site impurity system where impurity spins are located among host spins. The spin magnitude is s_0 for an impurity spin against s for a host spin. Exchange constants are J_0 for both sides of an impurity site and are J otherwise. The number of impurity spins in a block is $2h$. They are located on the sites of $\{k_j \mid j = 1, 2, \ldots, 2h\}$ in the block. Their distribution is random in the block and the impurity density is $\rho_0 = h/b$. In the calculation of $\tilde{s}_q$ on Eq. (11), we notice that it becomes $\tilde{s}_q^{(0)} \equiv \sum_{k=1}^{q}(-1)^{k+1}s$ for a pure spin system. We now replace the kth term in $\tilde{s}_q^{(0)}$ by $(-1)^{k+1}s_0$ if an impurity spin locates at the kth site. Then we have

$$\tilde{s}_q = \tilde{s}_q^{(0)} + (s - s_0)\sum_{j=1}^{2h}(-1)^{k_j}\theta(q - k_j), \qquad (22)$$

where the step function $\theta(x)$ is 1 for $x \geq 0$ and 0 otherwise. Taking the ensemble average with equal weight for all possible distributions, we have

$$\frac{2J^{(0)}}{J^{(1)}} = s\frac{1 + (1 + \frac{s_0}{s})\tilde{\rho}_0}{1 + 2\tilde{\rho}_0} \qquad (23)$$

with $\tilde{\rho}_0 = \rho_0(Js/J_0s_0 - 1)$. This is independent of the block size $2ba$ [15]. Derivation of this equation will be reported elsewhere. When $J_0s_0 < Js$, the gapless condition (21) gives the following results. In the case of $s = 1$, Eq. (21) has no integer solution of l for $s_0 \leq 2$. That is, impurities with $s_0 \leq 2$ do not force the gapful excitation of a homogeneous $s = 1$ spin system to be gapless [16,18]. In the case of $s = 1/2$, Eq. (21) has no integer solution of l for $1 \leq s_0 \leq 5/2$.

In summary, we obtained the NLSM (17) for a general antiferromagnetic Heisenberg spin chain with inhomogeneous spin magnitudes and inhomogeneous nearest-neighbor exchange constants arrayed in a finite period. We applied this formula to several cases and examined the gapless conditions. Since the formula is general, it can be applied to various cases which were not treated here.

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[15] The topological angle has the same value in each block and is therefore nonrandom in the continuum limit. A random topological angle with mean π would presumably produce a gap.
[16] The NLSM method does not apply to the case of $J_0/J \ll 1$ both in the bond impurity system and the site impurity system. The continuum approximation in the derivation of an NLSM is justified when adjacent spins are strongly coupled so that $\mathbf{n}_j$ changes slowly as a function of j. In the case of $J_0/J \ll 1$, however, local spin motion occurs about an impurity. For the $s = 1$ spin chain, an effective $s = 1/2$ spin appears based on the valence-bond-solid picture: I. Affleck, T. Kennedy, E. H. Lieb, and H. Tasaki, Phys. Rev. Lett. **59**, 799 (1987). Such $s = 1/2$ spins are observed: M. Hagiwara, K. Katsumata, I. Affleck, B. I. Halperin, and J. P. Renard, Phys. Rev. Lett. **65**, 3181 (1990); S. H. Glarum, S. Geshwind, K. M. Lee, M. L. Kaplan, and J. Michel, Phys. Rev. Lett. **67**, 1614 (1991).
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