were prepared from material supplied by Cominco American Inc., Spokane, Washington. The single crystal was prepared by a zone-leveling technique (from materials supplied by Johnson-Matthey & Co., Ltd., London, England) as described by C. Uher, H. J. Goldsmid, and J. R. Drabble, Phys. Status Solidi (b) 68, 709 (1975).

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Friedel-Type Oscillations in One-Dimensional Antiferromagnetic Insulators

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An impurity spin in one-dimensional spin-$\frac{1}{2}$ X-Y and antiferromagnetic Heisenberg models is shown to induce decaying spatial oscillations of magnetization for finite magnetic field. The wavelength of oscillation changes with magnetic field. Since these systems can be described mathematically in terms of a degenerate gas of pseudofermions, their response to impurity perturbation is similar to the response of an electron gas to a localized static charge.

It is known that the spin-$\frac{1}{2}$ X-Y and antiferromagnetic Heisenberg models with nearest-neighbor interaction in one dimension can be transformed into a problem of noninteracting (for X-Y) or interacting (for Heisenberg) fermions of zero chemical potential by using the Jordan-Wigner transformation.[1,2] This transformation has been used by many authors in the prediction and study of Peierls spin instability[3] in the compressible one-dimensional spin-$\frac{1}{2}$ X-Y model[4] and antiferromagnetic Heisenberg model,[5] and their Fermi-gas–like thermodynamic behavior.[6] In the present work this transformation is used to show the Friedel-type spatial oscillations of the pseudofermion density around an impurity in the one-dimensional spin-$\frac{1}{2}$ X-Y and antiferromagnetic Heisenberg models. The impurity magnetic atom has a spin-$\frac{1}{2}$ magnetic moment; however, it has an exchange integral J_1 with its two neighbors which differs from J, the nearest-neighbor exchange integral among the host spins. It is found that the magnetization induced at the ith site for large R_i is

$$\frac{\pi}{2}\left(\frac{J-J_1}{J}\right)\left(\frac{h}{J}\right)\frac{\sin(2k_F R_i)}{2k_F R_i},$$

where R_i is the distance of the ith site from the impurity, and k_F is a wave vector which depends on the ratio of the magnetic field h to the exchange integral J. This asymptotic oscillatory behavior disappears for zero magnetic field because of a particular momentum dependence of the pseudofermion scattering-matrix elements of the impurity perturbation. The physics behind this oscillation is the same as that behind the Friedel oscillation in a degenerate electron gas.[7] It arises because of the singular response of the Fermi gas in the ground state to perturbations with wave vector equal to twice the Fermi wave vector ($2k_F$).

Consider the case of the X-Y model. The Hamiltonian of the X-Y model containing an impurity at the origin is

$$H=J\sum_i(S_i^xS_{i+1}^x+S_i^yS_{i+1}^y)+h\sum_i S_i^z+(J_1-J)\sum_{\Delta=\pm1}(S_0^xS_\Delta^x+S_0^yS_\Delta^y), \tag{1}$$

where h is the magnetic field in energy units and S_i^x, S_i^y, S_i^z are the three components of the spin operator ($S=\frac{1}{2}$) at the ith site. By using the Jordan-Wigner transformation[1]

$$S_i^+\equiv S_i^x+iS_i^y=(2)^{i-1}\exp(i\pi\sum_{j<i}C_j^\dagger C_j)C_i^\dagger,\quad S_i^-\equiv S_i^x-iS_i^y=(2)^{i-1}C_i\exp(-i\pi\sum_{j<i}C_j^\dagger C_j),$$
$$S_i^z=\tfrac{1}{2}-C_i^\dagger C_i, \tag{2}$$

the Hamiltonian may be written as

$$H = J\sum_i C_i{}^\dagger C_i - h\sum_i C_i{}^\dagger C_i - \tfrac{1}{2}hN + \tfrac{1}{2}(J_1 - J)\sum_{\Delta=\pm 1}(C_0{}^\dagger C_\Delta + C_\Delta{}^\dagger C_0)$$

$$= \sum_k \epsilon_k C_k{}^\dagger C_k + \frac{J_1 - J}{N}\sum_k \{[\cos ka + \cos(k-q)a]C_k{}^\dagger C_{k-q}\} - \tfrac{1}{2}hN, \tag{3}$$

where

$$C_k{}^\dagger = \frac{1}{\sqrt{N}}\sum_n e^{ikR_n}C_n{}^\dagger, \quad C_k = \frac{1}{\sqrt{N}}\sum_n e^{-ikR_n}C_n,$$

$$\epsilon_k = J\cos ka - h.$$

Here $C_i{}^\dagger, C_i$ and $C_k{}^\dagger, C_k$ are the pseudofermion creation and annhilation operators, respectively, in the site and momentum representations, N is the total number of sites, and a is the lattice parameter. The chemical potential of these spinless fermions is zero.[2]

Consider the case in which the impurity perturbation is absent, i.e., $J = J_1$. The ground-state average of S_i^z in the absence of the magnetic field[1] is zero, i.e., $\langle S_i{}^z\rangle = 0$ for all i.[8] Using Eq. (2) we get $\langle C_i{}^\dagger C_i\rangle = \frac{1}{2}$. Thus the ground site corresponds to a half-filled band of noninteracting fermions. As we apply the magnetic field, the mean magnetization and hence the mean occupation number change. This means that the Fermi wave vector also changes with magnetic field. The Fermi wave vector is given by the solution of the equation[2,5]

$$\epsilon_{k_F} = J\cos k_F a - h = 0,$$

so that

$$k_F = a^{-1}\cos^{-1}(h/J). \tag{4}$$

The impurity spin only alters the exchange integral with its neighbors—this is a localized perturbation. So, now the problem reduces to studying the effect of a localized perturbation on a noninteracting Fermi gas. From the theory of degenerate Fermi system it is well known that the Fermi gas responds in a singular way at $2k_F$ in the ground state. In particular, the linear response in one dimension diverges logarithmically at $2k_F$. In linear-response theory, the induced fermion density or the induced magnetization $(\Delta\sigma_i)$ at the ith site is given by

$$\Delta\sigma_i = -\Delta n_i = -\frac{(J - J_1)}{N^2}\sum_k \frac{\cos ka(n_k - n_{k-q})}{\epsilon_k - \epsilon_{k-q}}\exp(iq - R_i). \tag{5}$$

where n_k is the Fermi distribution function. The factor $\cos ka$ arises from the momentum dependence of the one-body scattering potential, namely $[\cos ka + \cos(k-q)a]$ of Eq. (3). This particular dependence on the initial wave vector k and the momentum transfer $\hbar q$ gives rise to an interesting field dependence of the oscillatory part of $\Delta\sigma_i$. The above expression can be evaluated in the free-fermion approximation (i.e., using parabolic dispersion for the fermion energy) for large R_i as[9]

$$\Delta\sigma_i \approx \frac{\pi(J - J_1)}{2J}\frac{h}{J}\frac{\sin(2k_F R_i)}{2k_F R_i}. \tag{6}$$

Note that the above asymptotic oscillatory part of $\Delta\sigma_i$ vanishes identically as h tends to zero. This has the following meaning: The oscillatory behavior arises because of the logarithmic divergence of the response function at $2k_F$. The virtual transitions which give rise to this singular behavior correspond to a momentum transfer $\pm 2\hbar k_F$ and the initial momentum $\sim \pm hk_F$, respectively. At zero field, $k_F = \pi/2a$ and the matrix element of the above virtual transitions becomes very small and indeed vanishes identically for the initial wave vector of $\pm\pi/2a$ and momentum transfer of $\pm 2\hbar(\pi/2a)$: $\cos[(\pi/2a)a] + \cos[(\pi/2a - \pi/a)a] = 0$. At zero magnetic field the dominant asymptotic term behaves as $\sim 1/R_i^2$. As the magnetic field is changed, k_F moves away from $\pi/2a$ and the matrix element of the above processes become finite and the induced magnetization regains the asymptotic form (6). The free-fermion approximation is not valid for large magnetic field where $k_F \approx \pi/a$.

I have gone beyond linear-response theory and calculated the exact expression for the induced magnetization or fermion density in terms of the single-particle Green's functions[10-12] but I do not reproduce the results here.

Consider the pure Heisenberg antiferromagnetic system. Compared with the X-Y model this has the extra term $J\sum_i S_i^z S_{i+1}^z$. In terms of the pseudofermion operators, this term can be written as

$$-J\sum_k C_k^\dagger C_k + J\sum_{k,k'} \cos(qa) C_k^\dagger C_{k-q} C_{k'}^\dagger C_{k'+q}. \quad (7)$$

Bulaevskii[2] considered this antiferromagnetic model in the Hartree approximation and obtained the following results. The Hartree single-particle energy is

$$\epsilon_k = -2\sigma J - h + pJ\cos ka, \quad (8)$$

where

$$p = \tfrac{1}{2} - 2\sum_k \langle n_k\rangle \cos ka.$$

In the above expression, the average is taken in the noninteracting ground state with single-particle energy given by Eq. (8); p and σ are calculated in a self-consistent way. For $h/2J > 1$, there is complete ferromagnetic ordering; this corresponds to the completely filled band. Only the partially-filled-band case, where the impurity-induced magnetization oscillates, is of interest to us; this corresponds to $h/2J < 1$. For this latter case, the solution of the self-consistent equation is

$$h/J = [1 + (2/\pi)\cos\pi\sigma]\sin\pi\sigma + 2\sigma,$$

$$p = 1 + (2/\pi)\cos\pi\sigma. \quad (9)$$

The Fermi wave vector is given by the expression

$$\epsilon_{k_F} = -2\sigma J - h + pJ\cos k_F a = 0,$$

so that

$$k_F = \frac{1}{a}\cos^{-1}\left(\frac{h + 2\sigma J}{pJ}\right). \quad (10)$$

For $h/2J \lesssim \frac{1}{2}$, from Eq. (9) we find that $\sigma \approx h/J(\pi + 4)$ and hence

$$k_F \approx \frac{1}{a}\cos^{-1}\left\{\frac{h[1 + 2/(\pi+4)]}{J\{1 + (2/\pi)\cos[h\pi/(\pi+4)]\}}\right\}. \quad (11)$$

Thus the variation of k_F with h differs from that for the X-Y case.

The presence of the impurity gives additional one-body and two-body scattering terms,

$$(J_1 - J)\sum_{\Delta=\pm 1}(C_0^\dagger C_\Delta + C_\Delta^\dagger C_0) - \tfrac{1}{2}(J_1 - J)(C_{-1}^\dagger C_{-1} + 2C_0^\dagger C_0 + C_1^\dagger C_1) + (J_1 - J)\sum_{\Delta=\pm 1} C_0^\dagger C_0 C_\Delta^\dagger C_\Delta, \quad (12)$$

which can be treated in the Hartree approximation. After the Hartree approximation is made, the problem is similar to the X-Y problem and we get the following asymptotic expression for the induced magnetization:

$$\Delta\sigma_i \sim \pi\,\frac{(J - J_1)}{J}\,\frac{h[1 + 2/(\pi+4)]}{2J\{1 + (2/\pi)\cos[h\pi/(4+\pi)]\}}\,\frac{\sin(2k_F R_i)}{(2k_F R_i)}. \quad (13)$$

The amplitude of the oscillation vanishes for $h = 0$, again for the same reason elaborated in the X-Y case. The Hartree approximation discussed now predicts divergence of the susceptibility at $2k_F$ and hence the impurity-induced oscillation. Will this oscillation remain even if we go beyond the Hartree approximation? The susceptibility of an electron gas with short-range interaction in one dimension calculated by going beyond the Hartree approximation with use of renormalization-group techniques does show divergence at $2k_F$ in the ground state.[13] Since the present problem is mathematically similar to the electron-gas problem, one expects a similar divergence in the response function and hence the oscillation.

The oscillation discussed so far is a purely quantum mechanical phenomenon; it is easily shown to be absent in the classical ($S\to\infty$) X-Y and antiferromagnetic models in one dimension. So one expects it to happen for any finite impurity and host spins. A preliminary calculation by the author using the spin-wave approximation shows that there is oscillation for any finite-spin case.

In real systems there are three important factors which damp these oscillations. The first one is the temperature. The oscillation is expected only in highly degenerate Fermi gas and at temperature much lower compared with the Fermi temperature ($=pJ/k_B$), i.e., $k_B T/pJ \ll 1$. Around and above the Fermi temperature, there is only exponential damping and no oscillation. Much below the Fermi temperature and above the absolute zero of temperature, there is oscillation whose amplitude is at most damped by the term

$$\exp\left\{-\left(\frac{k_F R_i}{pJ/k_B T}\right)^2\right\}.$$

When $k_B T/pJ \sim 0.05$, this damping becomes effective only beyond a distance ten time the constant lattice from the impurity.

The second factor is the Peierls spin instability.[3-5] In the Peierls spin instability the spin-phonon (pseudofermion-phonon) interaction opens a gap at the Fermi surface below a critical temperature. This gap (Δ) also exponentially damps[14] the oscillatory term by

$$\exp\left\{\frac{-R_i k_F}{(pJ/\Delta)}\right\}.$$

The third factor is the three-dimensional magnetic ordering. The weak magnetic interaction among the chains becomes effective at very low temperatures and brings in three-dimensional magnetic ordering. Below the three-dimensional ordering, the present treatment is not valid and one intuitively expects strong damping. Hence the observations of these oscillations are possible at temperatures very low compared with the Fermi temperature but slightly above the largest of the Peierls instability temperature or the three-dimensional ordering temperature.

The compounds $CuCl_2 \cdot 5H_2O$ and $CuCl_2 \cdot 2NC_5H_5$ are good representatives[15] of one-dimensional spin-$\frac{1}{2}$ antiferromagnetic Heisenberg models. They also satisfy the above-mentioned requirements. The Fermi temperature ($=pJ/k_B$) and three-dimensional ordering temperature are 2.7°K and 0.03°K, respectively, for $CuCl_2 \cdot 5H_2O$; 20°K and 1.7°K, respectively, for $CuCl_2 \cdot 2NC_5H_5$. Copper which has been deprived of one of its d electrons is the magnetic ion in these compounds. The relatively abundant ^{63}Cu nucleus being a good NMR nucleus, Knight-shift studies by NMR[16] should reveal these oscillations at temperatures just above the three-dimensional ordering temperature. Moreover, the magnetic field necessary to have appreciable amplitude of oscillation (i.e., h/J) in these compounds is ~ 10 kG, which is not very high.

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