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SPIN RELAXATION IN CONDUCTING CHAINS

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Résumé. — Le temps de relaxation spin-réseau T_1 d'une chaîne de Hubbard est proportionnel, pour U grand, à la racine carrée de la fréquence de Larmor ω , dans le cas d'une bande non demi-remplie aussi bien que dans le cas (déjà connu) d'une bande demi-remplie. Le coefficient de proportionnalité a une limite finie pour $U \rightarrow \infty$, sauf pour une bande demi-remplie.

Abstract. — The spin lattice relaxation time T_1 of a Hubbard chain for large U is proportional to the square root of the Larmor frequency ω for a non-half-filled band as well as for the (already known) case of a half-filled band. The proportionality coefficient has a finite limit when U goes to ∞ , except for a half-filled band.

1. **Introduction.** — The measurement of the spin-lattice relaxation time T_1 in one-dimensional conductors by N.M.R. experiments is of special interest because the observed value of $1/T_1$ can be much greater than the Korringa value [1]. Measurements of $1/T_1$ as a function of the Larmor frequency ω actually exhibit a divergence ⁽¹⁾ when ω goes to 0. The agreement observed between certain experiments and an enhanced Korringa law [2] is probably limited to a small range of frequency and temperature ⁽¹⁾ [3].

A good approximate starting point for a theoretical treatment is provided by the Hubbard hamiltonian [4] :

$$\mathcal{H} = \theta \sum_{i=1}^N \sum_{\sigma=\uparrow,\downarrow} (c_{i\sigma}^+ c_{i+1,\sigma} + \text{h.c.}) + U \sum_i n_{i\uparrow} n_{i\downarrow} \quad (1)$$

$1/T_1$ is essentially proportional [4, 5, 6] to :

$$g(\omega) = \int_0^\infty \langle \mathbf{S}_i(0) \cdot \mathbf{S}_i(t) \rangle \cos \omega t \, dt \quad (2)$$

where :

$$S_i^+ = (S_i^-)^* = c_{i\uparrow}^+ c_{i\downarrow}, \quad S_i^z = \frac{1}{2}(c_{i\uparrow}^+ c_{i\uparrow} - c_{i\downarrow}^+ c_{i\downarrow}).$$

Before any general calculation of $g(\omega)$, it is of interest to discuss limiting cases :

i) For $U = 0$, $g(\omega)$ is proportional to $-\text{Log } \omega$ for $\omega \approx 0$, provided $T \neq 0$ [3].

ii) The case $U \gg \theta$, $\langle n_{i\uparrow} + n_{i\downarrow} \rangle = 1$ (half-filled band) is equivalent [4] to the Heisenberg model with $-J = \theta^2/U$, and it is found that [7]

$$g(\omega) \simeq (3 \hbar / 16 \theta) \sqrt{U/2 \hbar \omega}.$$

iii) A more difficult case is $U \gg \theta$, $U \gg T$ but $\langle n_{i\uparrow} + n_{i\downarrow} \rangle \neq 1$.

The present paper deals with case (iii). It will be shown that $g(\omega)$ again diverges like $1/\sqrt{\omega}$ for small ω . It will be assumed that

$$C \equiv \langle n_{i\uparrow} + n_{i\downarrow} \rangle < 1 \quad (3)$$

but the case $C > 1$ can be deduced from electron-hole symmetry.

2. **Formulation of the problem in terms of a probability distribution.** — In all calculations, the condition

$$U = \infty \quad (4)$$

will be assumed; the effect of a finite U will be briefly discussed in Section 5.

If (4) is satisfied, the spin σ_n of the n 'th electron is a constant of the motion. It follows that :

$$\langle S_i^z(0) S_i^z(t) \rangle = \sum_{n,m} \langle \sigma_n \sigma_m \rangle \langle \delta_{i,i_n(0)} \delta_{i,i_m(t)} \rangle \quad (5)$$

where $i_n(t)$ is the position of the n 'th electron at time t . Since there is no exchange for $U = \infty$,

$$\langle \sigma_n \sigma_m \rangle = \delta_{nm}/4 \quad (6)$$

⁽¹⁾ Devreux, F., private communication.

and insertion of (6) into (5) yields :

$$\langle S_i^z(0) S_i^z(t) \rangle = \frac{1}{4} CP(0, t) \quad (7)$$

where C is defined by (3) and $P(u, t)$ is the probability that the n 'th particle has moved u interatomic distances during the time t :

$$P(u, t) = \frac{1}{NC} \sum_i \langle \delta_{i, i_n(0)} \delta_{i+u, i_n(t)} \rangle. \quad (8)$$

3. Relation with density fluctuations. — The probability function $P(u, t)$ will now be expressed in terms of the density-density correlation function, by means of three assumptions which will be justified at the end of Section 4.

Instead of (8), the displacement $u(t)$ is the difference, $x(t) - x(0)$, of the positions of the n 'th electron at times t and 0, which are related to the density

$$\rho_i(t) = n_{i+}(t) + n_{i-}(t) \quad (9)$$

by the relation :

$$\sum_{i=x(0)}^{x(t)} \rho_i(t) = - \sum_{i=0}^{x(0)} [\rho_i(t) - \rho_i(0)]. \quad (10)$$

The first assumption is to replace $\rho_i(t)$ by its average value C on the left hand side of (10), yielding :

$$u(t) = - \frac{1}{C} \sum_{i=0}^{x(0)} [\rho_i(t) - \rho_i(0)]. \quad (11)$$

In principle, as shown in Section 4 below, for $U = \infty$, this formula permits the calculation of all moments :

$$\langle u^{2n}(t) \rangle = \sum_{u=0}^N u^{2n} P(u, t). \quad (12)$$

However, in order to avoid the calculation of complicated integrals, only the second moment ($n = 1$) will be explicitly calculated, so that $P(0, t)$ can only be obtained by means of a second assumption, namely a gaussian form for $P(u, t)$:

$$P(u, t) = P(0, t) \exp - u^2/\Delta^2. \quad (13)$$

Hence :

$$P(0, t) = [2\pi \langle u^2(t) \rangle]^{-1/2}. \quad (14)$$

4. Explicit calculation. — According to (2), (7), (14), (11), the calculation of T_1 reduces to that of the density-density correlation function. When (4) is satisfied, this correlation function is the same as for a one-dimensional, ideal gas of CN spinless fermions [8] described by the Hamiltonian :

$$\tilde{\mathcal{H}} = \sum_{i=1}^N (b_i^+ b_{i+1} + \text{h.c.}). \quad (15)$$

The density can be expressed in terms of the b_i 's as :

$$\rho_i = b_i^+ b_i. \quad (16)$$

The third assumption is that density correlations are independent of $x(0)$; now, eq. (11) and (16) yield :

$$\begin{aligned} \langle u^2(t) \rangle &= \frac{1}{C^2 N^2} \sum_{kk'} \langle b_k^+ b_k \rangle \times \\ &\times \langle b_{k'} b_{k'}^+ \rangle |1 - e^{i(\omega_k - \omega_{k'})t}|^2 \sum_{i=0}^{x(0)} \sum_{j=0}^{x(0)} e^{i(k-k')(i-j)}. \end{aligned}$$

For large $x(0)$, this expression is independent of $x(0)$, namely :

$$\begin{aligned} \langle u^2(t) \rangle &= \frac{2}{\pi^2 C^2} \int_{-\pi}^{\pi} dk \int_{-\pi}^{\pi} dk' \times \\ &\times f_k(1 - f_{k'}) \frac{\sin^2(\omega_k - \omega_{k'})t/2}{(k - k')^2} \quad (17) \end{aligned}$$

and this justifies the third assumption. We used :

$$f_k = \frac{1}{e^{\beta(\hbar\omega_k - \mu)} + 1}, \quad (18)$$

$$\hbar\omega_k = 2\theta(1 - \cos k) \quad (19)$$

and the chemical potential μ is defined by :

$$\frac{1}{2\pi} \int_{-\pi}^{\pi} f_k dk = C. \quad (20)$$

For small Larmor frequency ω , and $T \neq 0$, the dominant contribution to (2) is due to large t , as will be seen below; for large t , the dominant contribution to (17) comes from small values of $h = k' - k$, so that $\omega_{k'} - \omega_k$ can be replaced by $h d\omega_k/dk$; in addition, the integral over h can be extended from $-\infty$ to $+\infty$ and performed explicitly, yielding :

$$\langle u^2(t) \rangle = \frac{2t}{\pi C^2} \int_0^{\pi} f_k(1 - f_k) |d\omega_k/dk| dk. \quad (21)$$

This, together with (2), (7), (14), gives the result. In particular, at $T = \infty$ (actually, $\theta \ll k_B T \ll U$), $f_k = C$ and :

$$g(\omega) = \frac{3C}{16} \sqrt{\frac{C}{1-C}} \frac{\pi}{2} \frac{\hbar}{\theta\omega}. \quad (22)$$

At all non-vanishing T , $g(\omega)$ diverges like $\omega^{-1/2}$ at low frequency; the proportionality coefficient goes to infinity when T vanishes; at $T = 0$, it is directly seen from (17) that $\langle u^2(t) \rangle$ goes to a finite limit when t goes to infinity, and $g(\omega)$ reduces to $\delta(\omega)$, plus a probably regular function of ω . *Justification of the assumptions made in Section 3.*

i) The $(2n)$ th moment (12) can in principle be calculated from (11) and (16) : the highest order term in t is found to be $(2n-1)!! \langle u^2(t) \rangle^n$, and the gaussian law (13) results for large t if highest order terms in t^{n-1} , etc. are neglected.

ii) The mean square of the left hand side of (10) is well approximated by :

$$\sum_{i=0}^l \langle \rho_0 \rho_i \rangle = Cl^2 + \sum_{i=0}^l \langle \delta \rho_0 \delta \rho_i \rangle \quad (23)$$

where $l^2 = \langle u^2(t) \rangle$ and $\delta \rho_i = \rho_i - C$. The second term, easily obtained from (16), is proportional to l , and therefore negligible for large t . This justifies the approximation (11).

5. Intermediate cases. — 5.1 Finite U . We shall only discuss the high temperature case, where exchange effects are negligible; the situation can be depicted as a random motion of the spins, with a mean free path λ which goes to infinity when U vanishes. Since the velocity is of order $\theta/\hbar$, the mean square displacement of a spin is of order

$$\langle u^2(t) \rangle \approx \theta t / \lambda \hbar \quad (24)$$

and $g(\omega)$ is given by (2), (7), (14) and (24). Comparison with (22) shows that, if C and $(1 - C)$ are not too small, T_1 has the same order of magnitude (for given C, θ, ω) for all values of U except if U/θ is small. This is in agreement with experiments in quinolinium⁺ (TCNQ)₂⁻, where eq. (22) was found to be satisfied within experimental errors (¹).

5.2 Nearly half-filled band. For large U , and $C = 1$, as mentioned in the introduction, one has :

$$g(\omega) \simeq \frac{3 \hbar}{16 \theta} \sqrt{\frac{U}{2 \hbar \omega}}. \quad (25)$$

Comparison with (22) shows that (22) should hold if :

$$1 - C \gtrsim \sqrt{\theta/U}. \quad (26)$$

When $|1 - C|$ becomes smaller, T_1 is still proportional to $\sqrt{\omega}$, but the proportionality coefficient is greatly reduced.

6. Conclusion. — T_1 is proportional to $\sqrt{\omega}$ for a Hubbard chain at $T \neq 0$ in all cases except $U = 0$. Of course at low T [1], coupling with phonons and Peierls distortion can produce drastic effects which have not been considered here.

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