# Nuclear spin relaxation in a quasi-one-dimensional Hubbard band

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We have studied the effect of electron-electron and electron-impurity collisions on the frequency-dependent behavior of the nuclear spin relaxation rate in a one-dimensional conductor using the Hubbard model. The effect of occasional tunneling of an electron to adjacent chains is also examined. It is found that electron-electron collisions can seriously affect the electron spin diffusion in the nuclear spin relaxation even in systems where they have little effect on dc conduction. The limiting behavior of the present results is compared with the high-temperature and strong-coupling results obtained by previous authors.

## I. INTRODUCTION

The frequency-dependent nuclear spin relaxation is recognized as an important probe of the motion of spin carriers which interact with nuclear moments in solids. For example, the technique provides information about the dimensionality and degree of coherence of the motion of the carriers. Recent experimental results show evidence of one-dimensional diffusive motion of the mobile spins with a characteristic inverse square-root frequency dependence of the relaxation rate in organic conductors such as N-methyl-phenazinium (NMP)−tetracyanoquinodimethane (TCNQ) (Ref. 1), quinolinium-(TCNQ)$_2$ (Ref. 1), and tetrathiafulvalane (TTF)-TCNQ (Refs. 2 and 3) in the metallic regime, and in some excitonic systems[4] as well as in organic polymers such as undoped and heavily doped (i.e., metallic) polyacetylene (CH)$_x$ (Ref. 5) and metallic polyphenylene.[6]

As is well known, the nuclear spin relaxation in an ordinary metal is given by the Korringa rate.[7] The diffusive behavior in the metallic conductor cited above is a consequence of a short mean free path of the conduction electrons characteristic of low-dimensional metals. The purpose of this paper is to study the effect of electron-electron interaction, which is generally an important feature of the low-dimensional metals,[1,8,9] on the frequency-dependent behavior of the nuclear spin relaxation rate. The effect of impurity scattering will also be considered.

The nuclear spin relaxation rate is given by[1]

$$T_1^{-1} = \frac{1}{5\hbar^2}[6d^2\Phi^z(\omega_N)+(7d^2+5a^2)\Phi^+(\omega_e)] ,$$

(1.1)

where $d$ and $a$ are the dipolar and isotropic electron-proton hyperfine coupling constants, $\omega_N$ and $\omega_e$ are the nuclear and electronic Larmor frequencies, and[10]

$$\Phi^+(\omega)=2\hbar^{-2}\text{Re}\left[\int_0^\infty \langle s_i^+(t)s_i^-(0)\rangle e^{i\omega t}dt\right] ,$$

(1.2a)

$$\Phi^z(\omega)=2\hbar^{-2}\text{Re}\left[\int_0^\infty \langle s_i^z(t)s_i^z(0)\rangle e^{i\omega t}dt\right] .$$

(1.2b)

Here Re and angular brackets denote the real part and thermodynamic average. The quantities $s_i^z$, $s_i^+$, and $s_i^-$ represent, respectively, the $z$ component, the raising, and the lowering operators of the electronic spin at site $i$. In the present isotropic system one has

$$\Phi^+(\omega)=2\Phi^z(\omega) .$$

(1.3)

The nuclear and electronic Zeeman energies are much smaller than the thermal energy $k_BT$ and the Fermi energy.

The effect of isotropic impurity scattering has been considered by Fulde and Luther.[11] They found that the $\omega$ component of the spin correlation function exhibits a diffusive behavior when the electron relaxation rate $(\tau^{-1})$ becomes much larger than $\omega$. A generalized impurity scattering will be considered in this paper. Although the lifetime equals the transport relaxation time for isotropic scattering, it is expected that the time $\tau$ corresponds to the latter, a time scale beyond which the electron executes a random walk. For electron-electron scattering the situation is not as simple owing to the spin degree of freedom. We find that

for scattering between parallel spins $\tau^{-1}$ corresponds to the transport relaxation rate arising from $U$ processes. For antiparallel spins $\tau^{-1}$ is basically the decay rate of states at the Fermi level and arises from both $N$ and $U$ processes. This is due to the spin distinguishability of the particles involved in the collision. This fact has the following interesting consequence in a one-dimensional system. In such a metal even a large Coulomb interaction usually gives a vanishing contribution to dc resistivity because the resistive $U$ process is allowed only for a half-filled band. On the other hand, it gives an important effect on the electron spin diffusion in the nuclear spin relaxation problem.

The effect of Coulomb scattering on the nuclear spin relaxation rate has been studied in a Hubbard chain by Devreux[1] in the high-temperature strong-coupling limit $k_B T >> 4t_z$, $U \geq t_z$ by using a short time approximation for the correlation function, and by Villain[12] in the $U = \infty$ limit in terms of a spinless fermion model. Here $t_z$ and $U$ are the nearest-neighbor tunneling integral and on-site Coulomb repulsion, respectively. Although limiting behaviors of a system are of interest, the above assumptions are made for mathematical tractability of the problem rather than for practical reasons.

In this paper we study the spin correlation function using the Boltzmann equation derived by a microscopic theory. The present treatment includes both weak- and strong-coupling situations, except that for the latter a low density is assumed. This method allows for a general treatment of the effect of Coulomb scattering. A specific form of the model is used later to obtain an explicit result. For this purpose we use the Hubbard chain model. In the strong-coupling limit, our result agrees with that of Ref. 12 but disagrees with that of Ref. 1, as will be discussed in Sec. IV. The effect of interchain hopping as well as the combined effect of general static and Coulomb collisions is examined. The present result is not valid in the insulator regime where the band is half-filled and $U$ is larger than the bandwidth. This case is equivalent to the Heisenberg model and has been treated earlier.[13,14]

In the next section the formalism is given and the model is defined. In Sec. III the basic transport equation is established for the spin correlation function. The frequency-dependent behavior of the nuclear spin relaxation rate is examined in detail for the impure Hubbard chain model. The effect of interchain tunneling is studied. The results are summarized in Sec. IV and discussions are given.

## II. FORMALISM

The system is described by the Hamiltonian

$$H = \sum_{\vec{k}\sigma} \epsilon_{\vec{k}\sigma} a^{\dagger}_{\vec{k}\sigma} a_{\vec{k}\sigma} + \sum_{j} \sum_{\vec{k}\vec{k}'\sigma} \exp[i(\vec{k}'-\vec{k})\cdot\vec{R}_j] I_{\vec{k}'\vec{k}} a^{\dagger}_{\vec{k}'\sigma} a_{\vec{k}\sigma}$$
$$+ \frac{1}{2} \sum_{\vec{k}\vec{k}'\sigma} \sum_{\vec{k}_1\vec{k}'_1\sigma_1} \langle \vec{k}'\sigma, \vec{k}'_1\sigma_1 | V | \vec{k}\sigma, \vec{k}_1\sigma_1 \rangle a^{\dagger}_{\vec{k}'\sigma} a^{\dagger}_{\vec{k}'_1\sigma_1} a_{\vec{k}_1\sigma_1} a_{\vec{k}\sigma} , \tag{2.1}$$

where $\epsilon_{\vec{k}\sigma}$, $\vec{R}_j$, $a^{\dagger}_{\vec{k}\sigma}$, and $a_{\vec{k}\sigma}$ are, respectively, the Bloch energy of an electron of wave vector $\vec{k}$ and spin $\sigma$, the impurity position, and the creation and annihilation operators. The second term in (2.1) describes impurity scattering and the third term the electron-electron interaction. The Hubbard model corresponds to

$$\epsilon_{\vec{k}\sigma} = \sum_{\alpha=x,y,z} 2t_\alpha(1 - \cos k_\alpha a_\alpha) \tag{2.2}$$

and

$$\langle \vec{k}'\sigma, \vec{k}'_1\sigma_1 | V | \vec{k}\sigma, \vec{k}_1\sigma_1 \rangle$$
$$= U\Delta(\vec{k}+\vec{k}_1, \vec{k}'+\vec{k}'_1)\delta_{\sigma,-\sigma_1} . \tag{2.3}$$

Namely, contact interaction is allowed only for op-

posite spins. The quantities $t_\alpha$, $a_\alpha$, and $\delta$ are the nearest-neighbor tunneling integral, lattice constant, and Kronecker's delta, respectively. The symbol $\Delta$ signifies conservation of wave vectors. The normalization constants for wave-vector sums will be omitted.

In order to evaluate the spin correlation function microscopically, it is convenient to rewrite (1.2) as

$$\Phi^z(\omega) = 2 \sum_{\vec{q}} \text{Im} \frac{F_{\vec{q}}(\hbar\omega + i0) - F_{\vec{q}}(i0)}{\omega} , \tag{2.4}$$

where Im denotes imaginary part and $F_{\vec{q}}(\hbar\omega + i0)$ is the analytic continuation to slightly above the real axis of the dimensionless quantity,
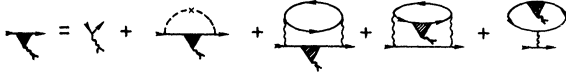
FIG. 1. Integral equation for the vertex part.



FIG. 2. Electronic self-energy part.

$$F_{\vec{q}}(\hbar\omega_r)=\hbar^{-2}\beta^{-1}\int_0^\beta \langle \hat{T}s_{\vec{q}}^z(u)s_{\vec{q}}^z(0)\rangle e^{\hbar\omega_r u}du \ ,$$

(2.5)

defined on the imaginary axis $\hbar\omega_r=2\pi k_B T r i$. Here $r$ is an integer, $\beta^{-1}=k_B T$, and $s_q^z(u)$ is in the imaginary time Heisenberg representation. The quantity $\hat{T}$ is a "time"-ordering operator and

$$s_{\vec{q}}^z=\hbar\sum_{\vec{k}} \tfrac{1}{2}(a_{\vec{k}+\vec{q},\uparrow}^\dagger a_{\vec{k},\uparrow}-a_{\vec{k}+\vec{q},\downarrow}^\dagger a_{\vec{k},\downarrow}) \ .$$

(2.6)

The basic equation for the spin-density fluctuation function $\Phi^z(\omega)$ is the Boltzmann equation given in (3.2). Although this form of transport equation is expected from the Fermi-liquid theory, we derive it here microscopically to include strong-coupling and nondegenerate situations. The transport equation is obtained by evaluating the spin correlation function in (2.5) in terms of Feynman diagrams. Its structure is similar to that of the velocity correlation function responsible for the conductivity tensor recently studied for the same system.[15] Therefore analogous details of the calculation will be referred to this work. Only basic approximations and results will be given here.

The correlation function $F_{\vec{q}}(\hbar\omega_r)$ is obtained by joining the two horizontal full-electron propagators (solid lines) entering and exiting from the vertex part (shaded triangl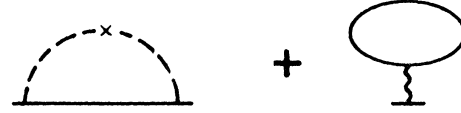e) on the left-hand side of the vertex equation shown in Fig. 1 into a loop. The first term on the right-hand side of the vertex equation describes an unperturbed system. The second term represents the single impurity scattering (dashed line) and the last three terms the dressed electron-electron scattering (curvy lines) for which a $T$-matrix approximation is used. For a weak interaction the latter reduces to the Born approximation. For a strong interaction it describes multiple scattering at a low electron concentration. Only scattering between antiparallel spins will be considered. The effect of scattering between parallel spins will be discussed later qualitatively. The wiggly external incoming lines for the vertex part denote input of momentum $\hbar\vec{q}$ and energy $\hbar\omega_r$.

The electron self-energy part is given in Fig. 2, and the $T$ matrix is defined in Fig. 3 in terms of bare scattering (dotted lines), introduced in (2.3). The present approximation is valid in the regime where the mean free path is larger than the de Broglie wavelength.[15] The perturbation to the electronic spectrum is assumed to be small. The evaluation of the integral equations in Figs. 1 and 3 has essentially been worked out in Ref. 15. In the next section the final results will be given in terms of a transport equation. The nuclear spin relaxation will be evaluated by solving the transport equation.

## III. NUCLEAR SPIN RELAXATION RATE

The basic results of the evaluation of the spin correlation function and the vertex equation introduced in Fig. 1 are summarized by the following equations[15]:

$$\Phi^z(\omega)=\frac{1}{\beta\omega}\sum_{\vec{q}}\sum_{\vec{k}}(-f'_{\vec{k}})\mathrm{Re}\phi_{\vec{k}} \ ,$$

(3.1)

and

$$-i(\omega-\omega_{\vec{k},\vec{q}})\phi_{\vec{k}}=\omega_{\vec{k},\vec{q}}-\frac{2\pi}{\hbar}N_i\sum_{\vec{k}'}|I_{\vec{k}'\vec{k}}|^2(\phi_{\vec{k}}-\phi_{\vec{k}'})\delta(\epsilon_{\vec{k}}-\epsilon_{\vec{k}'})$$

$$+\beta(f'_k)^{-1}\sum_{k',k_1,k'_1}[\phi_{\vec{k}}+(-\phi_{\vec{k}_1})-\phi_{\vec{k}'}-(-\phi_{\vec{k}'_1})]P_{\vec{k}\,\vec{k}_1}^{\vec{k}'\,\vec{k}'_1} \ ,$$

(3.2)

where $\omega_{\vec{k},\vec{q}}=\hbar^{-1}(\epsilon_{\vec{k}+\vec{q}}-\epsilon_{\vec{k}})$, $N_i$ is the total number of impurities, $f_{\vec{k}}$ the Fermi function, and $f'_{\vec{k}}$ its derivative with respect to $\epsilon_{\vec{k}}$. The distribution function $\phi_k$ corresponds to the vertex part in Fig. 1. The scattering-in terms in (3.2) arise from the vertex correction represented by the last four diagrams in Fig. 1, while the scattering-out terms come from the electronic self-energy part in Fig. 2. The quantities $\phi_k$ in (3.2) are independent of spin and dependent on $\vec{q}$ and $\omega$. The first, second, and third terms on the right-hand

side of (3.2) describe driving term, impurity, and Coulomb collisions, respectively. Without the last two terms, (3.2) describes a noninteracting system, yielding the Korringa rate for the nuclear spin relaxation.

The quantity $P_{\vec{k}\,\vec{k}_1}^{\vec{k}',\,\vec{k}_1'}$ describes the multiple scattering rate of two particles of opposite spins from an initial state of wave vectors $\vec{k}$ and $\vec{k}_1$ into a final state $\vec{k}'$ and $\vec{k}_1'$, respectively:

$$P_{\vec{k}\,\vec{k}_1}^{\vec{k}'\,\vec{k}_1'}=\frac{2\pi}{\hbar}\,|\,K(\vec{k},\vec{k}_1;\vec{k}',\vec{k}_1')\,|^2 f_{\vec{k}}f_{\vec{k}_1}(1-f_{\vec{k}'})(1-f_{\vec{k}_1'})\,\delta(\epsilon_{\vec{k}}+\epsilon_{\vec{k}_1}-\epsilon_{\vec{k}'}-\epsilon_{\vec{k}_1'})\Delta(\vec{k}+\vec{k}_1,\vec{k}'+\vec{k}_1')\;.\tag{3.3}$$

For the on-site contact interaction in (2.3) the $T$ matrix (cf. Fig. 3) reads[15]

$$K(\vec{k},\vec{k}_1;\vec{k}',\vec{k}_1')=\frac{U}{1-UL}\;,\tag{3.4}$$

where

$$L=\sum_{\vec{q}}\frac{1-f_{\vec{k}-\vec{q}}-f_{\vec{k}_1+\vec{q}}}{\epsilon_{\vec{k}}+\epsilon_{\vec{k}_1}-\epsilon_{\vec{k}-\vec{q}}-\epsilon_{\vec{k}_1+\vec{q}}-i0}\;.\tag{3.5}$$

The inversion of signs for the quantites $\phi_{\vec{k}_1}$ and $\phi_{\vec{k}_1'}$ in the Coulomb scattering term of (3.2) is due to the fact that for antiparallel spins the product $\langle\uparrow|\sigma^z|\uparrow\rangle\langle\downarrow|\sigma^z|\downarrow\rangle$ in the spin correlation function of (2.5) becomes negative. On the other hand, for collision between parallel spins the signs of the quantities $\phi_{\vec{k}_1}$ and $\phi_{\vec{k}_1'}$ in (3.2) should be reversed as in the regular Boltzmann equation.

## A. Hubbard chain

### 1. Transport equation

In a one-dimensional system the energy delta function in (3.3) has a simple form:

$$\delta(\epsilon_k+\epsilon_{k_1}-\epsilon_{k'}-\epsilon_{k_1'})=\delta\left[4t\cos\frac{k+k_1}{2}\sin\frac{k-k'}{2}\sin\frac{k_1-k'}{2}\right]\;.\tag{3.6}$$

The directional indices are dropped and the lattice constant is assumed to be unity. The resonances at $k+k_1=\pm\pi$ (corresponding to $U$ and $N$ processes) of the delta function in (3.6) are unimportant because the $T$ matrix vanishes there. The resonance at $k_1=k'$ corresponds to exchange of momenta (i.e., $k'=k_1$ and $k_1'=k$), yielding,

$$\delta(\epsilon_{k_1}+\epsilon_{k_1}-\epsilon_{k_1}-\epsilon_{k'})=\frac{1}{t\,|\,\sin k-\sin k'\,|}\delta(k_1-k')\;.$$

The transport equation in (3.2) is then rewritten as

$$-i(\omega-\omega_{k,q})\phi_k=\omega_{k,q}-(2\tau_{ik})^{-1}(\phi_k-\phi_{-k})-\frac{2}{\hbar\beta t}\sum_{k'}|\,K(k,k';k',k)\,|^2(-f_{k'}')\frac{\phi_k-\phi_{k'}}{|\,\sin k-\sin k'\,|}\;,\tag{3.7}$$

where the impurity-scattering transport relaxation rate

$$\tau_{ik}^{-1}=\frac{2\pi}{\hbar}\rho(\epsilon_k)N_i\,|\,I_{-k,k}\,|^2\tag{3.8}$$

arises from transitions across the Fermi sea, and $\rho(\epsilon_k)$ is the density of states per spin. Note that the factor $\phi_k-\phi_{k_1}-\phi_{k'}+\phi_{k_1'}$ in (3.2) does not vanish for the momentum-exchange $N$ processes owing to the sign re-

versal of the $\phi_{k_1}$ and $\phi_{k_1'}$ terms for antiparallel spins. For parallel spins this factor will be of the form $\phi_k$ + $\phi_{k_1} - \phi_{k'} - \phi_{k_1'}$ and nonvanishing only for $U$ processes just as in the dc transport phenomenon. The $T$ matrix in (3.7) is given in the low-density limit by

$$|K(k,k';k',k)|^2 = \frac{U^2}{1 + \dfrac{U^2}{4t^2(\sin k - \sin k')^2}} .$$ (3.9)

## 2. Low-temperature behavior

At low temperatures $(k_B T \ll \epsilon_F$, $\epsilon_F$ is the Fermi energy) one has $-f'_{k'} = \delta(\epsilon_{k'} - \epsilon_F)$ and (3.7) reduces to

$$-i(\omega - \omega_{k,q})\phi_k = \omega_{k,q} - (2\tau_{ik})^{-1}(\phi_k - \phi_{-k})$$

$$-\frac{\rho_F}{\hbar\beta t}\left[\frac{|K(k,k_F;k_F,k)|^2}{|\sin k - \sin k_F|}(\phi_k - \phi_{k_F}) + \frac{|K(k,-k_F;-k_F,k)|^2}{|\sin k + \sin k_F|}(\phi_k - \phi_{-k_F})\right] ,$$ (3.10)

where $\rho_F = \rho(\epsilon_F)$ and $k_F$ is the Fermi wave number. One now multiplies (3.10) by $-f'_k$ and sums over $k$ in the vicinity of $k = k_F$, obtaining

$$[-i(\omega - \omega_{k_F,q}) + (2\tau^*)^{-1}]\phi_{k_F}$$

$$-(2\tau^*)^{-1}\phi_{-k_F} = \omega_{k_F,q} .$$ (3.11a)

The $k$ summation in the first term in the large parentheses on the right-hand side of (3.10) is cancelled out. Note also from (3.9) that $K(k,k;k,k)$ $= 0$. Similarly, it follows for $k = -k_F$ that

$$-(2\tau^*)^{-1}\phi_{k_F} + [-i(\omega - \omega_{-k_F,q})$$

$$+ (2\tau^*)^{-1}]\phi_{-k_F} = \omega_{-k_F,q} .$$ (3.11b)

In (3.11) $\tau_i = \tau_{ik_F}$ and

$$\tau^{*-1} = \tau_i^{-1} + \tau_U^{-1} ,$$ (3.12)

with

$$\tau_U^{-1} = \frac{|K(k_F,-k_F;-k_F,k_F)|^2\rho_F}{\hbar\beta t \sin k_F} .$$ (3.13)

Note that while $\tau_i$ is the transport relaxation time due to impurity scattering, $\tau_U$ is basically the lifetime due to electron-electron scattering. The physical meaning of the latter was discussed earlier.
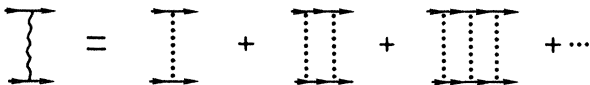


FIG. 3. $T$ matrix approximations.

The $T$ matrix is given by

$$|K(k_F,-k_F;-k_F,k_F)|^2 = \frac{U^2}{1 + \dfrac{2U^2}{\epsilon_F(4t - \epsilon_F)}} .$$ (3.14)

In the limit $\omega\tau^* \ll 1$, important contributions to $\Phi^z(\omega)$ come from $q \ll l^{-1}$ where $l = v_F\tau^*$ is the mean free path. Approximating

$$\omega_{k,q} = v_k q ,$$ (3.15)

where $v_k = (1/\hbar)(d\epsilon_k/dk)$, one finds from (3.11)

$$\phi_{\pm k_F} = \frac{-iDq^2 \mp ilq\omega}{Dq^2 - i\omega} .$$ (3.16)

Here $D = \tau^* v_F^2$ is the diffusion constant. Inserting (3.16) in (3.1), one finds

$$\Phi^z(\omega) = \frac{k_B T\rho_F}{2\sqrt{2D\omega}} .$$ (3.17)

Contributions from $q \geq l^{-1}$ including $q = \pm 2k_F$ are smaller than that in (3.17) by a factor $(2\omega\tau^*)^{1/2}$.

In the limit $\omega\tau^* \gg 1$, the vertex correction is not necessary. Setting $\tau^{*-1} = 0$ in (3.11), one finds

$$\phi_{\pm k} = \frac{i\omega}{\omega - \omega_{\pm k,q} + i0} .$$ (3.18)

The poles $q = \pm\omega/v_F$ and $q = \pm 2k_F$ of $\Phi_{\pm k_F}$ contribute equally to $\Phi^z$, yielding the Korringa result

$$\Phi^z(\omega) = \frac{k_B T\rho_F}{v_F} = \hbar\pi k_B T\rho_F^2 .$$ (3.19)

This expression is independent of the frequency $\omega$.

### 3. Approximate general behavior

At low temperatures the dominant contribution to the Coulomb scattering in (3.7) is shown to arise from "horizontal" transitions across the Fermi sea. The "vertical" transition in the neighborhood of $k' \simeq k$ is less important even at higher temperatures due to the cancellation of the factor $\Phi_k - \Phi_{k'}$ in the $k'$ summation as well as due to the vanishing of the $T$ matrix at $k' = k$. Noting that the summand in (3.7) is a smoothly behaving function of $k'$, we approximate [cf. (3.9)]

$$\frac{|K(k,k';k',k)|^2}{|\sin k - \sin k'|} \simeq \frac{U^2\lambda^{-1}}{1+U^2/4t^2\lambda^2} = U_{\text{eff}}^2 .$$

(3.20)

The magnitude of $\lambda$ becomes $\lambda = 2\sin k_F$ at low temperatures and roughly $\lambda \sim 2$ at high temperatures (i.e., $k_B T \gg 4t$). Although crude, this approximation makes the transport equation (3.7) soluble without distorting the underlying physics.

The transport equation (3.7) is then simplified as

$$-i(\omega - \omega_{k,q})\Phi_k = \omega_{k,q} - (2\tau_{ik})^{-1}(\phi_k - \phi_{-k})$$
$$-\frac{2U_{\text{eff}}^2}{\hbar\beta t}\sum_{k'}(-f'_{k'})(\phi_k - \phi_{k'}) .$$

(3.21)

Defining

$$\tau_U^{*-1} = \frac{2U_{\text{eff}}^2}{\hbar\beta t}\sum_k (-f'_k)$$

(3.22)

and

$$\psi = \sum_k (-f'_k)\phi_k ,$$

(3.23)

one rewrites (3.21) as

$$[-i(\omega - \omega_{k,q}) + \tfrac{1}{2}(\tau_k^{*-1} + \tau_U^{*-1})]\phi_k$$
$$= \omega_{k,q} + \tfrac{1}{2}\tau_{ik}^{-1}\phi_{-k} + \tau_U^{*-1}\psi \bigg/ \bigg[-\sum_{k'} f'_{k'}\bigg] ,$$

(3.24a)

and similarly,

$$[-i(\omega - \omega_{-k,q}) + \tfrac{1}{2}(\tau_k^{*-1} + \tau_U^{*-1})]\phi_{-k}$$
$$= \omega_{-k,q} + \tfrac{1}{2}\tau_{ik}^{-1}\phi_k + \tau_U^{*-1}\psi \bigg/ \bigg[-\sum_{k'} f'_{k'}\bigg] ,$$

(3.24b)

where

$$\tau_k^{*-1} = \tau_{ik}^{-1} + \tau_U^{*-1} .$$

(3.25)

In the limit $\omega\tau_k^* \gg 1$, (3.24) reduces to the Korringa result (3.18). In the opposite limit $\omega\tau_k^* \ll 1$ important contributions arise from $q \ll l_k^{-1}$ where $l_k = v_k\tau_k^*$. In this limit one finds from (3.24), using (3.15) and the self-consistency condition (3.23),

$$\psi = \frac{-i\tau_U^*\sum_k(-f'_k)\tau_k^*v_k^2q^2}{1-g\bigg/\sum_k(-f'_k)} ,$$

(3.26)

where

$$g = \tau_U^{*-1}\sum_k (-f'_k)\frac{-i\omega + \tau_k^{*-1}}{-\omega^2 + v_k^2q^2 - i\omega(\tau_k^{*-1} + \tau_U^{*-1}) + \tau_U^{*-1}\tau_k^{*-1}} .$$

(3.27)

The summand in (3.27) is expanded in terms of $\omega\tau^*$ and $ql_k$, yielding

$$g = \sum_k (-f'_k)[1 - \tau_U^*(\tau_k^*v_k^2q^2 - i\omega)]$$

(3.28)

and

$$\psi = \frac{-iDq^2\sum_k(-f'_k)}{Dq^2 - i\omega} ,$$

(3.29)

where

$$D = \langle\tau_k^*v_k^2\rangle \equiv \sum_k(-f'_k)\tau_k^*v_k^2 \bigg/ \sum_k(-f'_k) .$$

(3.30)

Finally, one finds from (3.1) and (3.29)

$$\Phi^z(\omega) = \frac{k_B T\sum_k(-f'_k)}{2\sqrt{2D\omega}} .$$

(3.31)

This expression reduces to (3.17) at low temperatures.

## B. Interchain Diffusion

The linear chain model considered in Sec. III A is highly idealized. In an actual system the electron can eventually tunnel to a neighboring chain, given enough time (namely, for a small Larmor frequency). In the following we study the effect of a small (i.e., $t_x << k_B T, t_z$) interchain tunneling. For

example, the ratio of the interchain to interchain tunneling integrals for TTF-TCNQ is of order $t_x/t_z \sim 10^{-3}$. For simplicity we ignore the impurity scattering (i.e., $\tau_{ik} = \infty$) and consider the low-temperature $(k_B T << \epsilon_F)$ diffusion limit $(\omega \tau^* << 1)$.

The intrachain Coulomb scattering is still described in the manner given in Sec. III A 1. In analogy with (3.7), one has $(t_y = 0)$

$$-i(\omega - \vec{v}_{\vec{k}} \cdot \vec{q})\phi_k(k_x) = \vec{v}_{\vec{k}} \cdot \vec{q} - \frac{1}{\hbar \beta t} \sum_{k'} \frac{|K(k,k';k',k)|^2}{|\sin k - \sin k'|}(-f'_{k'})[\phi_k(k_x) - 2\phi_{k'} + \phi_k], \qquad (3.32)$$

where $v_{\vec{k}}^\alpha = 2 t_\alpha a_\alpha \hbar^{-1} \sin k_\alpha a_\alpha$, $k \equiv k_z$, and

$$\phi_k = \sum_{k_x} \phi_k(k_x). \qquad (3.33)$$

The $T$ matrix is independent of the transverse wave vectors $k_x$.

Defining the lifetime of the state $k$ by

$$\tau_k^{0-1} = \frac{1}{\hbar \beta t} \sum_{k'} \frac{|K(k,k';k',k)|^2}{|\sin k - \sin k'|}(-f'_{k'}), \qquad (3.34)$$

and

$$p_n(k)^{-1} = \sum_{k_x} \frac{(\vec{v}_{\vec{k}} \cdot \vec{q})^n}{-i(\omega - \vec{v}_{\vec{k}} \cdot \vec{q}) + \tau_k^{0-1}} \qquad (n = 0,1) \quad (3.35)$$

one finds from (3.32) and (3.33)

$$[p_0(k_F) + \tau_0^{-1}]\phi_{k_F} - 2\tau_0^{-1}\phi_{-k_F}$$
$$= p_0(k_F)p_1(k_F)^{-1}, \qquad (3.36a)$$

and similarly

$$-2\tau_0^{-1}\phi_k + [p_0(-k_F) + \tau_0^{-1}]\phi_{-k_F}$$
$$= p_0(-k_F)p_1(-k_F)^{-1}, \qquad (3.36b)$$

where

$$\tau_0^{-1} = \tau_{k_F}^{0-1} = \frac{|K(k_F, -k_F; -k_F, k_F)|^2}{4\hbar \beta t \sin k_F} \rho_F. \qquad (3.34')$$

The coupled equations (3.36) are solved. Expanding

$$p_1(\pm k_F)^{-1} \simeq -i\tau_0 D_x q_x^2 \pm v_F q_z \tau_0 - i v_F^2 q_z^2 \tau_0^2, \qquad (3.37)$$

and

$$p_0(\pm k_F) \simeq \tau_0^{-1}[1 - i\tau_0(\omega \mp v_F q_z) + \tau_0 D_x q_x^2], \qquad (3.38)$$

one finds

$$\phi_{\pm k_F} = \frac{-i \sum_\alpha D_\alpha q_\alpha^2 \mp \frac{1}{4} i \tau_0 \omega v_F q_z}{\sum_\alpha D_\alpha q_\alpha^2 - i\omega}. \qquad (3.39)$$

In (3.39) the intrachain diffusion constant is given by $D_z = \tau^* v_F^2$ and

$$D_\alpha = \tau_0 \sum_{k_\alpha} (v_{k_\alpha}^\alpha)^2 \equiv \tau_0 \langle (v_{k_\alpha}^\alpha)^2 \rangle \quad (\alpha \neq z). \qquad (3.40)$$

The quantity in (3.40) describes the interchain diffusion constant of an electron which is in a band motion along the chain and occasionally tunnels to the adjacent chain. The expression in (3.40) agrees with that derived earlier.[15] Note that the interchain diffusion constant is proportional to the life time $\tau_0$ while $D_z$ is proportional to $\tau^*$. This is due to the fact that the duration time for interchain tunneling is limited by the lifetime of the state in the chain. This is more clearly seen by rewriting (3.40) as[15]

$$D_\alpha = \sum_{k_\alpha} (\tau_0 v_{k_\alpha}^\alpha)^2 / \tau_0.$$

The numerator represents the transition probability during time $\tau_0$ and summand the transition rate from each mode.

Finally, inserting (3.39) in (3.1), one obtains

$$\Phi^z(\omega) = \beta^{-1} \rho_F \sum_g \frac{\sum_\alpha D_\alpha q_\alpha^2}{\left[\sum_\alpha D_\alpha q_\alpha^2\right]^2 + \omega^2}, \qquad (3.41)$$

a well-known form in the diffusion problem. The detailed frequency-dependent behavior of $\Phi^z(\epsilon)$ can be obtained by a numerical integration. In the limit $\omega_z >> \omega >> \omega_x, \omega_y$ where $\omega_\alpha = D_\alpha a_\alpha^{-2}$, the expression in (3.41) reduces to the one-dimensional result (3.17). A two-dimensional behavior $\Phi^z(\omega)\alpha \ln\omega$ is obtained for $\omega_x, \omega_z >> \omega >> \omega_y$, while three-dimensional behavior $\Phi^z(w)\alpha\omega^{1/2}$ is expected for $\omega_x, \omega_y, \omega_z >> \omega$.

## IV. SUMMARY AND DISCUSSIONS

The frequency-dependent behavior of the nuclear spin relaxation rate is studied in a quasi-one-dimensional Hubbard band. An explicit expression is obtained for the spin correlation function both in the diffusion limit $(\omega\tau^* << 1)$ and Korringa regime $(\omega\tau^* >> 1)$. The combined effect of impurity scattering and electron-electron interaction is examined. The effect of interchain tunneling is investigated. The present result is valid outside the insulating regime where the Coulomb correlation is strong and the band is nearly half-filled.

In the following we study various limiting behaviors of the present results in the diffusion limit $(\omega\tau^* << 1)$ and compare them with the high-temperature $(k_B T >> 4t)$ strong-coupling $(U >> 4t)$ results obtained previously[1,12] for a Hubbard chain. Defining $n$ as the number of electrons per site, we find from (3.22), (3.30), and (3.31),[16]

$$\Phi^z(\omega) = \frac{n(1-n/2)}{4\sqrt{2D\omega}} , \qquad (4.1)$$

$$D = \frac{2t^2 a^2}{\hbar^2}\tau^* , \qquad (4.2)$$

$$\tau^{*-1} = \frac{4t\lambda}{\hbar}n(1-n/2) . \qquad (4.3a)$$

The above result is valid at low concentration $(n << 1)$. If one sets $\lambda \simeq 2$, the expression in (4.1) agrees with that of Villain[12] to within a factor-of-order unity. However, it disagrees with that of Devreux[1] who found $D \propto n^{-1/2}$. Such a concentration dependence is difficult to understand because, as noted in the present work, the diffusion constant is linear in the scattering time $\tau^*$ which is expected to be inversely proportional to the number of scatterers (i.e., $n$). In quinolinium-(TCNQ)$_2$, for example, one has[1] $t = 0.04 - 0.05$ eV, $U = 0.15$ $-0.18$ eV, and $n = 0.5$, and the diffusive behavior is obtained for $\omega < \tau^{*-1} = 2.4 \times 10^{14}$ sec$^{-1}$.

In the high-temperature weak-coupling limit

$(U << 4t)$ the results in (4.1) and (4.2) are valid for all concentrations, and the collision rate is given by

$$\tau^{*-1} = \frac{U^2\lambda^{-1}}{\hbar t}n(1-n/2) , \qquad (4.3b)$$

yielding

$$\Phi^z(\omega) = \frac{U}{8t}\left[\frac{\hbar}{\omega ta\lambda}\right]^{1/2}[n(1-n/2)]^{3/2} . \qquad (4.4)$$

Note that all the quantities in (4.1)—(4.4) are independent of the temperature.

At low temperatures the quantity $\Phi^z(\omega)$ is found from (3.12), (3.13), and (3.17):

$$\Phi^z(\omega) = \frac{k_B T\rho_F}{2v_F\sqrt{2\omega}}(\tau_i^{-1}+A\hbar^{-1}k_B T)^{1/2} , \qquad (4.5)$$

where $A$ is a dimensionless constant

$$A = 2\pi |K(k_F, -k_F; -k_F, k_F)|^2\rho_F^2 . \qquad (4.6)$$

In (4.5) the temperature dependence is shown explicitly and the contribution $\tau_i^{-1}$ from impurity scattering is included. In a one-dimensional metal electron-electron collisions are nonresistive for dc transport because the momentum-dissipating $U$ processes are not allowed, in general, except for a half-filled band. In the present problem, however, due to the distinguishability of spins involved in the collision between antiparallel spins, even the $N$ processes dissipate the momentum of a given spin; from the point of view of an up spin, a down spin represents a moving impurity which provides an inelastic scattering potential. Therefore, the diffusion constant obtained from a nuclear spin relaxation measurement does not necessarily determine the dc electron mean free path when electron-electron collisions are involved. This effect should be important in one-dimensional organic conductors[1-3] and metallic polymers[5,6] and reveals itself through the characteristic temperature dependence in (4.5).[17]

The second term in the parentheses of (4.5) represents the Coulomb scattering rate $\tau_U^{-1}$ defined in (3.13) and equals

$$\tau_U^{-1} \simeq 3.4 \times 10^{11} T\left|\frac{K}{4t}\right|^2\frac{1}{\sin^2 k_F} \text{ (sec}^{-1}) ,$$

where $T$ is in units of K. For a filling of 0.6 electron per site of the conducting TCNQ chain and for $K/4t \sim 1/2$, one has

$$\tau_U^{-1} \simeq 1.3 T \times 10^{11} \text{ (sec}^{-1})$$

This value is comparable to the transport relaxation rate (expressed in $sec^{-1}$) in TTF-TCNQ deduced from the dc metallic resistivity data[18] '(80—300 K):

$$\tau_{tr}^{-1} \simeq 1.76T \times 10^{11} \ (\text{sec}^{-1}) \ .$$

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