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The Korringa Relation for the Impurity Nuclear Spin-Lattice Relaxation in Dilute Kondo Alloys

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We present a proof of the Korringa relation for the impurity nuclear spin-lattice relaxation of transition-metal impurities in simple metals. Two important mechanisms are considered; the relaxation due to the d-spin fluctuations and the d-orbital fluctuations. By using the 5-fold degenerate Anderson model it is shown that the Korringa relation is valid at low temperatures $T \ll T_K$ for these two mechanisms in any order of the Coulomb interaction between d-electrons. The proof can be easily extended to the Korringa relation for the Wolff model.

§ 1. Introduction

The electronic structure of transition-metal impurities in simple metals is well described by the Anderson model.[1] A merit of this model is that it allows us a systematic study of dilute alloys containing transition elements. Exact theories of the Anderson model are desired for this reason.

It is believed that the ground state of the Anderson model is a nonmagnetic singlet for any finite values of the Coulomb repulsion U on the impurity orbital. Since the ground state should be a smooth function of U, it is quite natural then to expect that the ground state for any finite U can be obtained from the one for $U=0$ by the perturbation expansion. Recently Yamada and Yosida[2] have developed a theory in this direction, and studied in detail low-temperature properties*) of the symmetric single-band Hamiltonian (i.e., the impurity d-orbital is half-filled). A remarkable point of this theory is that they have found some exact relations among the low-temperature impurity contributions of the specific heat, the magnetic susceptibility and the resistivity. The relations remain valid irrespective of the magnitude of U as far as the perturbation expansion in U converges. When U is very large (i.e., the s-d limit), the relations Yamada and Yosida found coincide with what Nozières[3] has discovered on the basis of the s-d model and a phenomenological Fermi-liquid theory. One may regard therefore the Yamada-Yosida theory as a sort of microscopic justification of the Fermi-liquid description of dilute Kondo alloys.

*) We mean by low temperature the region $T \ll T_K$ (Kondo temperature), where T_K is defined as the inverse of the static impurity susceptibility.

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The purpose of this paper is to apply the Yamada and Yosida's approach to *dynamical response functions* and to give a proof of the Korringa relation (KR) for the impurity nuclear spin-lattice relaxation[4] at low temperatures $T \ll T_K$ and low fields $H \ll H_K (H_K \sim k_B T_K / \mu_B)$. Two mechanisms are believed to be important to the nuclear spin-lattice relaxation of transition-metal impurities; the hyperfine coupling of the impurity nuclear spin with the d-spin fluctuations (core polarization) and with the orbital moment fluctuations. The KR between the nuclear spin-lattice relaxation time T_1 and the Knight shift K is stated as[4]

$$\begin{cases} K_{(d)}^2 T_{1(d)} T = C_{(d)} , \\ K_{(\mathrm{orb})}^2 T_{1(\mathrm{orb})} T = C_{(\mathrm{orb})} , \end{cases} \tag{1·1}$$

where the suffixes d and orb indicate the d-spin and d-orbital moment mechanisms, respectively. $C_{(d)}$ and $C_{(\mathrm{orb})}$ are constants. $T_{1(d)}^{-1}$ ($T_{1(\mathrm{orb})}^{-1}$) is proportional to the imaginary part of the low-frequency spin (orbital) susceptibility, while $K_{(d)}$ ($K_{(\mathrm{orb})}$) is essentially the static spin (orbital paramagnetic) susceptibility:[4]

$$\begin{cases} K_{(d)} = A_{(d)} \lim_{\omega \to 0} \mathrm{Re}\, \chi_{\mathrm{spin}}^{zz} (\omega + i\delta), \\ T_{1(d)}^{-1} = k_B T A_{(d)}^2 (g_n \mu_n)^2 \lim_{\omega \to 0} \left[\frac{1}{\omega} \mathrm{Im} (\chi_{\mathrm{spin}}^{+-} (\omega + i\delta)) \right] \end{cases} \tag{1·2a}$$

and

$$\begin{cases} K_{(\mathrm{orb})} = A_{(\mathrm{orb})} \lim_{\omega \to 0} \mathrm{Re}\, \chi_{\mathrm{orb}}^{zz} (\omega + i\delta), \\ T_{1(\mathrm{orb})}^{-1} = k_B T A_{(\mathrm{orb})}^2 (g_n \mu_n)^2 \lim_{\omega \to 0} \left[\frac{1}{\omega} \mathrm{Im} (\chi_{\mathrm{orb}}^{+-} (\omega + i\delta)) \right], \end{cases} \tag{1·2b}$$

where $A_{(d)}$ and $A_{(\mathrm{orb})}$ represent the hyperfine coupling constants, and g_n and μ_n are the g-factor of the impurity nucleus and the nuclear Bohr magneton, respectively. $\chi_{\mathrm{spin}}(\omega + i\delta)$ and $\chi_{\mathrm{orb}}(\omega + i\delta)$ are the frequency-dependent spin and orbital paramagnetic susceptibilities of the impurity d-electrons, respectively:

$$\begin{cases} \chi_{\mathrm{spin}}^{zz} (\omega + i\delta) = (2\mu_B)^2 \int_0^\infty dt e^{i(\omega + i\delta)t} i \langle [S_d^z(t),\ S_d^z(0)] \rangle \\ \qquad \equiv (2\mu_B)^2 \langle\langle S_d^z ;\ S_d^z \rangle\rangle_{\omega + i\delta} , \\ \chi_{\mathrm{orb}}^{zz} (\omega + i\delta) = \mu_B^2 \int_0^\infty dt e^{i(\omega + i\delta)t} i \langle [L^z(t),\ L^z(0)] \rangle \\ \qquad \equiv \mu_B^2 \langle\langle L^z ;\ L^z \rangle\rangle_{\omega + i\delta} \end{cases} \tag{1·3}$$

with the total d-spin S_d^z and the total orbital angular momentum L^z. For a small field $H \ll H_K$ there is no distinction between the longitudinal and transverse susceptibilities; $\chi^{+-}(\omega + i\delta) = 2\chi^{zz}(\omega + i\delta)$ holds. Hence (1·1) is equivalent to

$$\lim_{\omega \to 0} \left[\frac{1}{\omega} \mathrm{Im}\, \chi_{\mathrm{spin}} (\omega + i\delta) \right] = [\chi_{\mathrm{spin}}(0)]^2 / C_{(d)} k_B (g_n \mu_n)^2 \tag{1·4a}$$

and

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$$\lim_{\omega\to 0}\left[\frac{1}{\omega}\,\mathrm{Im}\,\chi_{\mathrm{orb}}(\omega+i\delta)\right]=[\chi_{\mathrm{orb}}(0)]^2/C_{(\mathrm{orb})}k_B(g_n\mu_n)^2\,. \tag{1·4b}$$

Here we have used χ instead of χ^{zz} since the direction of the oscillating magnetic field is irrelevant. Both sides of (1·4) should be evaluated at zero temperature. We prove this relation quite generally on the basis of the Anderson model with 5-fold degeneracy[5),6)]

$$H=\sum_{k\sigma}\varepsilon_k a^+_{k\sigma}a_{k\sigma}+\sum_{km\sigma}(V_{km}a^+_{k\sigma}d_{m\sigma}+\mathrm{h.c.})$$
$$+\sum_{m\sigma}E_d d^+_{m\sigma}d_{m\sigma}+\sum_{\substack{mm'\\ \sigma\sigma'}}(Ud^+_{m\sigma}d^+_{m'\sigma'}d_{m'\sigma'}d_{m\sigma}+Jd^+_{m\sigma}d^+_{m'\sigma'}d_{m\sigma'}d_{m'\sigma}), \tag{1·5}$$

where $a_{k\sigma}$ denotes the creation operator of conduction electrons and $d_{m\sigma}$ is that of the impurity d-electron with the orbital angular momentum m. ε_k and E_d are measured from the chemical potential. The s-d mixing V_{km} is taken as $V_{km}=V\cdot\sqrt{4\pi}Y_{lm}(\Omega_k)$ with the spherical harmonics $Y_{lm}(\Omega_k)$. The last term describes the Coulomb and exchange interaction on the d-orbitals. The Hamiltonian (1·5) has rotational invariance in the spin and real space,[5)] and is suitable for the present purpose.

The KR (1·4) has been proved by Dworin and Narath[5)] within the *random phase approximation* (RPA), which starts from the nonmagnetic Hartree-Fock (HF) ground state. According to the RPA $C_{(d)}$ and $C_{(\mathrm{orb})}$ are given by

$$\begin{cases} C_{(d)}=(2l+1)\,(2\mu_B/g_n\mu_n)^2/2\pi k_B\,, \\ C_{(\mathrm{orb})}=\dfrac{4}{3}l(l+1)\,(2l+1)\,(\mu_B/g_n\mu_n)^2/2\pi k_B\,. \end{cases} \tag{1·6}$$

The HF approximation predicts a discontinuous transition from the nonmagnetic to the magnetic state with the increase of Coulomb interaction, which is an artifact inherent in the HF. Higher order effects neglected in the HF theory wash it out completely and lead to a smooth "transition". That is to say, the exact $\chi(\omega+i\delta)$ for a relatively large Coulomb interaction is greatly different from that in the HF-RPA. Therefore one may expect a deviation from (1·4a) (1·4b) and (1·6). In spite of this, as we show, the KR remains valid beyond the HF-RPA as far as the perturbation expansion in terms of U and J is convergent.

The paper is arranged as follows: Section 2 is devoted to the first step of proof, which is based on the Friedel sum rule. In § 3 we make an analysis of the low-frequency spin susceptibility and prove the KR for the d-spin fluctuation mechanism. The proof of the KR for the orbital fluctuation mechanism is presented in § 4. Some related problems are discussed in the last section.

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§ 2. Friedel sum rule and impurity spin susceptibility at zero temperature

The Friedel sum rule for the Anderson model was first proved by Langreth,[7] using the perturbation expansion for the single-band Anderson model at zero temperature. Following Langreth, we prove the Friedel sum rule with a slight generalization in order to discuss later the impurity spin susceptibility.

First we add a small spin-dependent term $H'_{\rm spin}$ to the d-electron part of (1·5):

$$H'_{\rm spin} = -\sum_{m\sigma} \delta\mu_{d\sigma} d^+_{m\sigma} d_{m\sigma} . \tag{2·1}$$

We study the Friedel sum rule for the *combined* Hamiltonian $H+H'_{\rm spin}$. The total localized charge $\Delta n_{d\sigma}$ with spin σ is given by

$$\Delta n_{d\sigma} = \sum_m n_{dm\sigma} + \sum_k (n_{k\sigma} - n^0_{k\sigma}) , \tag{2·2}$$

where $n_{dm\sigma}$ and $n_{k\sigma}$ are the d-electron and conduction-electron numbers, respectively. $n^0_{k\sigma}$ represents $n_{k\sigma}$ for the pure host metal. These quantities can be expressed with the complete d-electron Green's function $G_{ddm\sigma}(\omega+i\delta)$, the conduction-electron one $G_{kk\sigma}(\omega+i\delta)$ and the Green's function for the pure host $G^0_{k\sigma}(\omega+i\delta)$. (2·2) is then equivalent to

$$\begin{aligned}\Delta n_{d\sigma} = & \int_{-\infty}^{\infty} d\varepsilon f(\varepsilon)\Big(-\frac{1}{\pi}\,{\rm Im}\Big)\Big[\sum_m G_{ddm\sigma}(\varepsilon+i\delta) \\ & + \sum_k (G_{kk\sigma}(\varepsilon+i\delta) - G^0_{k\sigma}(\varepsilon+i\delta))\Big] \\ = & \int_{-\infty}^{\infty} d\varepsilon f(\varepsilon)\Big(-\frac{1}{\pi}\,{\rm Im}\Big)\sum_m G_{ddm\sigma}(\varepsilon+i\delta)\,(1+|V|^2\sum_k G^0_{k\sigma}(\varepsilon+i\delta)) .\end{aligned} \tag{2·3}$$

Here use has been made of the relation

$$\begin{aligned}G_{kk\sigma}(\varepsilon+i\delta) &= G^0_{k\sigma}(\varepsilon+i\delta) + [G^0_{k\sigma}(\varepsilon+i\delta)]^2 \sum_m V_{km} G_{ddm\sigma}(\varepsilon+i\delta) V_{mk} \\ &= G^0_{k\sigma}(\varepsilon+i\delta) + [G^0_{k\sigma}(\varepsilon+i\delta)]^2 |V|^2 \sum_m G_{ddm\sigma}(\varepsilon+i\delta) .\end{aligned} \tag{2·4}$$

$f(\varepsilon)$ represents the Fermi distribution function. Introducing the proper self-energy part $\Sigma_{dm\sigma}(\varepsilon+i\delta)$ by

$$\begin{aligned}G_{ddm\sigma}(\varepsilon+i\delta) &= [\varepsilon - E_{d\sigma} - |V|^2 \sum_k G^0_{k\sigma}(\varepsilon+i\delta) - \Sigma_{dm\sigma}(\varepsilon+i\delta)]^{-1} , \\ E_{d\sigma} &\equiv E_d - \delta\mu_{d\sigma} .\end{aligned} \tag{2·5}$$

(2·3) can be transformed to*)

$$\Delta n_{d\sigma} = \int_{-\infty}^{\infty} d\varepsilon f(\varepsilon)\Big(-\frac{1}{\pi}\,{\rm Im}\Big)\sum_m \Big[\frac{\partial}{\partial\varepsilon} \ln (E_{d\sigma} + |V|^2 \sum_k G_{k\sigma}(\varepsilon+i\delta)$$

*) As a matter of fact $G_{ddm\sigma}$ and $\Sigma_{dm\sigma}$ are independent of m because all the m's are equivalent.

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$$+\Sigma_{dm\sigma}(\varepsilon+i\delta)-\varepsilon)+G_{ddm\sigma}(\varepsilon+i\delta)\frac{\partial}{\partial\varepsilon}\Sigma_{dm\sigma}(\varepsilon+i\delta)\Big]. \qquad (2\cdot6)$$

A key point is that at $T=0°\mathrm{K}$,

$$\int_{-\infty}^{\infty}d\varepsilon f(\varepsilon)\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_m\left[G_{ddm\sigma}(\varepsilon+i\delta)\frac{\partial}{\partial\varepsilon}\Sigma_{dm\sigma}(\varepsilon+i\delta)\right]=0 \qquad (2\cdot7)$$

holds for $H+H'_{\mathrm{spin}}$. An analogous relation was first pointed out by Luttinger[8] for the *homogeneous* Fermi liquid. However the relation has nothing to do with the momentum conservation in the homogeneous system, but only the energy conservation plays an important role. According to Luttinger the relation (2·7) for a *fixed* σ is satisfied as far as the interaction between electrons is spin independent. In the J term of (1·4) only orbital exchanges occur. Therefore the electron-electron interaction in (1·4) is spin independent and (2·7) holds. From (2·6) we obtain at $T=0°\mathrm{K}$

$$\Delta n_{d\sigma}=\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_m \ln(E_{d\sigma}-i\Delta+\Sigma_{dm\sigma}(i\delta)). \qquad (2\cdot8)$$

For the constant density of states of conduction electrons

$$|V|^2\sum_k G_{k\sigma}(i\delta)=-i\pi|V|^2\rho=-i\Delta\,. \qquad (2\cdot9)$$

This has been used in (2·8). Defining the phase shift at the Fermi energy by*)

$$\delta_\sigma=\frac{\pi}{2}-\arctan\frac{E_{d\sigma}+\Sigma_{dm\sigma}(i\delta)}{\Delta}\,, \qquad (2\cdot10)$$

we find from (2·8)

$$\Delta n_{d\sigma}=(2l+1)\frac{1}{\pi}\delta_\sigma\,, \qquad (2\cdot11)$$

which is nothing but the Friedel sum rule.

Now we take the derivative of (2·8) with respect to $\delta\mu_{d\sigma'}$. The result

$$\left.\frac{\partial\Delta n_{d\sigma}}{\partial\delta\mu_{d\sigma'}}\right|_{\delta\mu=0}=\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_m\left[\frac{\delta_{\sigma\sigma'}-(\partial\,\Sigma_{dm\sigma}(i\delta)/\partial\delta\mu_{d\sigma'})|_{\delta\mu=0}}{-E_d-\Sigma_{dm\sigma}(i\delta)+i\Delta}\right] \qquad (2\cdot12)$$

is very useful. The left-hand side of (2·12) is a response of the localized charge to a small change of the impurity chemical potential. It is related to the fluctuation of the d-electron numbers:**)

$$\left.\frac{\partial\Delta n_{d\sigma}}{\partial\delta\mu_{d\sigma'}}\right|_{\delta\mu=0}=\int_0^\beta d\tau\langle\tilde{n}_{d\sigma}(\tau)\tilde{n}_{d\sigma'}(0)\rangle$$

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*) $\Sigma_{dm\sigma}(i\delta)$ describes the many-body effect at the Fermi energy, and is a real quantity. This is one of the Fermi-liquid properties of the present model.

**) Actually the localized charge $\Delta n_{d\sigma}$ contains the d-electron part $\sum_m n_{dm\sigma}$ as well as the conduction-electron part $\sum_k(n_{k\sigma}-n^0_{k\sigma})$ (See (2·2)). The latter contribution however vanishes for the constant density of states of conduction electrons (i.e., the d-electron density of states for $U=J=0$ is exactly Lorentzian). This is Anderson's compensation theorem.[1]

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$$=\langle\langle \tilde{n}_{d\sigma};\ \tilde{n}_{d\sigma'}\rangle\rangle_{i0} \tag{2·13}$$

with $\tilde{n}_{d\sigma}(\tau)\equiv e^{H\tau}\tilde{n}_{d\sigma}e^{-H\tau}$ and $\tilde{n}_{d\sigma}\equiv\sum_m(d^+_{m\sigma}d_{m\sigma}-\langle d^+_{m\sigma}d_{m\sigma}\rangle)$. The average $\langle\cdots\rangle$ is taken with (1·5). Combining (2·13) with (2·12) we obtain

$$\langle\langle \tilde{n}_{d\sigma};\ \tilde{n}_{d\sigma'}\rangle\rangle_{i0}=\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_m\left[\frac{\delta_{\sigma\sigma'}-(\partial\,\Sigma_{dm\sigma}/\partial\delta\mu_{d\sigma'})\,|_{\delta\mu=0}}{-E_d-\Sigma_{dm\sigma}(i\delta)+i\Delta}\right], \tag{2·14}$$

which leads to the following expression for the static impurity spin susceptibility at zero temperature:

$$\begin{aligned}\chi_{\mathrm{spin}}(0)/(2\mu_B)^2 &=\langle\langle S_d^z;\ S_d^z\rangle\rangle_{i0}\\ &=\frac{1}{4}\sum_{\sigma\sigma'}\sigma\sigma'\langle\langle \tilde{n}_{d\sigma};\ \tilde{n}_{d\sigma'}\rangle\rangle_{i0}\\ &=\left(-\frac{1}{2\pi}\,\mathrm{Im}\right)\sum_m\left[\frac{1-((\partial\Sigma_{dm\uparrow}(i\delta)/\partial\delta\mu_{d\uparrow})-(\partial\Sigma_{dm\uparrow}(i\delta)/\partial\delta\mu_{d\downarrow}))\,|_{\delta\mu=0}}{-E_d-\Sigma_{dm\sigma}(i\delta)+i\Delta}\right].\end{aligned} \tag{2·15}$$

This relation is substituted into the right-hand side of (1·4a). In the next section the left-hand side of (1·4a) is discussed.

§ 3. Low-frequency spin susceptibility at zero temperature

Since the impurity spin is quenched at low temperatures $T\ll T_{K\,\mathrm{spin}}$,*) the imaginary part of the dynamical spin susceptibility is proportional to the frequency ω at low frequencies $\omega\ll T_{K\,\mathrm{spin}}$.

We make an analysis of the dynamical susceptibility with the thermodynamic Green's function defined as

$$\begin{aligned}\int_0^\beta d\tau e^{i\omega\tau}\langle T[S_d^z(\tau)S_d^z(0)]\rangle &\equiv\langle\langle S_d^z;\ S_d^z\rangle\rangle_{i\omega}\\ &=\tfrac{1}{4}\sum_{\sigma\sigma'}\sigma\sigma'\langle\langle \tilde{n}_{d\sigma};\ \tilde{n}_{d\sigma'}\rangle\rangle_{i\omega},\qquad \omega=2\pi n/\beta\,.\ \ (n:\ \text{integer})\end{aligned} \tag{3·1}$$

The retarded Green's function (1·3) can be obtained by the analytic continuation $i\omega\to\omega+i\delta$. Especially the ω-linear imaginary part of $\langle\langle \tilde{n}_{d\sigma};\ \tilde{n}_{d\sigma'}\rangle\rangle_{\omega+i\delta}$ is obtained from the $|\omega|$-linear contribution of $\langle\langle \tilde{n}_{d\sigma};\ \tilde{n}_{d\sigma'}\rangle\rangle_{i\omega}$ at small ω. It is because $|\omega|=(-i\,\mathrm{sgn}\,\omega)i\omega$ goes over to $(-i)\omega$ by the analytic continuation. In the zero temperature limit the discrete energies in the thermodynamic Green's function formal-

*) $T_{K\,\mathrm{spin}}$ is the Kondo temperature for the impurity spin, which is most conveniently defined as the inverse of $\chi_{\mathrm{spin}}(0)$ at zero temperature. We can also define $T_{K\,\mathrm{orb}}$, the Kondo temperature for the orbital moment quenching as the inverse of $\chi_{\mathrm{orb}}(0)$ at zero temperature. It is expected that $T_{K\,\mathrm{spin}}<T_{K\,\mathrm{orb}}$ in general.

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ism are replaced by corresponding continuous ones. Our analysis hereafter is devided into 3 steps:*)

[1] We apply to $\langle\langle\tilde{n}_{d\sigma};\ \tilde{n}_{d\sigma'}\rangle\rangle_{i\omega}$ the perturbation expansion in terms of U and J and consider a contribution of a certain order of U and J, using the Feynman diagram. Each diagram consists of d-electron lines and interaction vertices; the former represents the unperturbed d-electron Green's function $G^0_{dm\sigma}(i\varepsilon)=[i\varepsilon-E_d+i\Delta\,\mathrm{sgn}\,\varepsilon]^{-1}$, while U or J is attached to the latter. In order to pick up the $|\omega|$-linear contribution from a given diagram, we leave the ω-linear contribution only in one of the d-electron lines. For the other d-electron lines the limit $\omega\to 0$ is taken carefully, since an extra factor $i\cdot\mathrm{sgn}\,\omega$ may appear from the singularity of $G^0_{dm\sigma}(i\varepsilon)$ at $\varepsilon=0$.

A typical diagram of $\langle\langle\tilde{n}_{d\sigma};\ \tilde{n}_{d\sigma'}\rangle\rangle_{i\omega}$ looks like Fig. 1. Here the diagram has been divided into two parts at an ω-dependent d-electron line: The left part Γ_1 has an external vertex with incoming energy ω, while the right part Γ_2 contains a vertex with outgoing energy ω. These two parts are connected with each other by an even number of d-electron lines, one of which has ω-dependence in it. This division of the diagram is arbitrary as far as the left and right parts have only one external vertex. It is easy to see that the $|\omega|$-linear contribution is obtained only when the total number of d-electron lines at the division is two. The proof for this goes as follows: The ω-linear contribution of Fig. 1 has the form

$$-\sum_{m,m'}\sum_{\substack{m_1\cdots m_{2n}\\ \sigma_1\cdots\sigma_{2n}}}\int_{-\infty}^{\infty}\cdots\int\frac{d\varepsilon_1\cdots d\varepsilon_{2n}}{(2\pi)^{2n}}\Gamma_1{}^{(\sigma\sigma_1\cdots\sigma_{2n})}(m;\ m_1i\varepsilon_1,\ \cdots,\ m_{2n}i\varepsilon_{2n})$$

$$\times\Gamma_2{}^{(\sigma'\sigma_1\cdots\sigma_{2n})}(m';\ m_1-i\varepsilon_1,\ \cdots,\ m_{2n}-i\varepsilon_{2n})$$

$$\times i\omega\frac{dG^0_{dm_1\sigma_1}(i\varepsilon_1)}{di\varepsilon_1}G^0_{dm_2\sigma_2}(i\varepsilon_2)\cdots G^0_{dm_{2n}\sigma_{2n}}(i\varepsilon_{2n})2\pi\delta\Big(\sum_{j=1}^{2n}\varepsilon_j\Big).$$

Substituting the relation

$$\frac{d}{di\varepsilon}G^0_{dm\sigma}(i\varepsilon)=-[G^0_{dm\sigma}(i\varepsilon)]^2+2\,\mathrm{Im}[G^0_{dm\sigma}(+i0)]\delta(\varepsilon)$$

into the above expression, we find that the integrand is not singular if $n\geqq 2$; after the integrations the above expression does not contain any $i\cdot\mathrm{sgn}\ \omega$ factor in it. For $n=1$ the singularities of $G^0_{dm_1\sigma_1}(i\omega+i\varepsilon_1)$ and $G^0_{dm_1\sigma_1}(i\varepsilon_1)$ overlap in the limit $\omega\to 0$. Figure 2 shows the general structure of diagrams with $n=1$. A close examination of $G^0_{dm_1\sigma_1}(i\omega+i\varepsilon_1)G^0_{dm_1\sigma_1}(i\varepsilon_1)$ in Fig. 2 gives us the $|\omega|$-linear term $-2(i\omega)(i\cdot\mathrm{sgn}\ \omega)[\mathrm{Im}\ G^0_{dm_1\sigma_1}(+i0)]^2\delta(\varepsilon_1)=2|\omega|\ [\mathrm{Im}\ G^0_{dm_1\sigma_1}(+i0)]^2\ \delta(\varepsilon_1)$, which is exactly what we need.

[2] Next we consider the effect of the self-energy insertion to $G^0_{dm_1\sigma_1}(i\omega+i\varepsilon_1)\cdot$

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*) Throughout this proof we follow essentially Yamada and Yosida's analysis[2] of Green's functions by extending it to degenerate orbitals with an arbitrary d-electron number.

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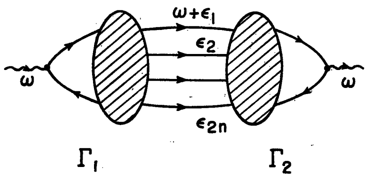

Fig. 1. The structure of a diagram of $\langle\langle \tilde{n}_{d\sigma};\tilde{n}_{d\sigma'}\rangle\rangle_{i\omega}$ for a given order of U and J. It has two vertex parts Γ_1 and Γ_2 and an even number of d-electron lines connecting Γ_1 with Γ_2.

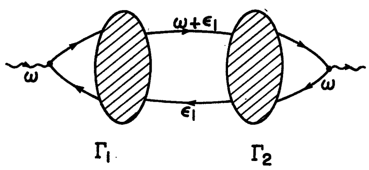

Fig. 2. The structure of diagrams, which give nonvanishing $|\omega|$-linear contributions in the small ω limit.

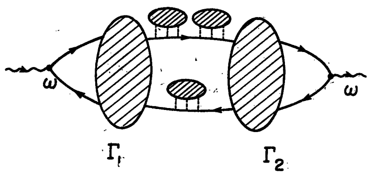

Fig. 3. The self-energy insertion to the two d-electron propagators, which connect Γ_1 with Γ_2.

$G^0_{dm_1\sigma_1}(i\varepsilon_1)$, which is shown in Fig. 3. This type of contribution is expressed as

$$-\sum_{m,m'}\sum_{m_1\sigma_1}\int_{-\infty}^{\infty}\frac{d\varepsilon_1}{2\pi}\Gamma_1{}^{(\sigma\sigma_1\sigma_1)}(m;\ m_1i\varepsilon_1,\ m_1-i\varepsilon_1)\Gamma_2{}^{(\sigma'\sigma_1\sigma_1)}(m';\ m_1i\varepsilon_1,\ m_1-i\varepsilon_1)$$
$$\times G^0_{dm_1\sigma_1}(i\omega+i\varepsilon_1)\,[1-\Sigma_{dm_1\sigma_1}(i\omega+i\varepsilon_1)G^0_{dm_1\sigma_1}(i\omega+i\varepsilon_1)]^{-1}$$
$$\times G^0_{dm_1\sigma_1}(i\varepsilon_1)\,[1-\Sigma_{dm_1\sigma_1}(i\varepsilon_1)G^0_{dm_1\sigma_1}(i\varepsilon_1)]^{-1}. \tag{3·2}$$

The ω-dependence of $\Sigma_{dm_1\sigma_1}(i\omega+i\varepsilon_1)$ does not give us any $|\omega|$-linear contribution; therefore the self-energy insertion does not change the analysis in [1] except for the replacement of $2|\omega|[\mathrm{Im}\,G^0_{dm\sigma}(+i0)]^2$ by $2|\omega|\,[\mathrm{Im}\,G_{ddm\sigma}(+i0)]^2$, where $G_{ddm\sigma}(i\varepsilon)$ is the complete d-electron Green's function

$$G_{ddm\sigma}(i\varepsilon)=[i\varepsilon-E_d+i\Delta\ \mathrm{sgn}\ \varepsilon-\Sigma_{dm\sigma}(i\varepsilon)]^{-1}.$$

The $|\omega|$-linear contribution of (3·2) is then given by

$$-\frac{|\omega|}{\pi}\sum_{mm'}\sum_{m_1}[\mathrm{Im}\,(G_{ddm\sigma}(+i0)]^2\sum_{\sigma_1}\Gamma_1{}^{(\sigma\sigma_1\sigma_1)}(m;\ m_1i0,\ m_1i0)$$
$$\times\Gamma_2{}^{(\sigma'\sigma_1\sigma_1)}(m';\ m_1i0,\ m_1i0). \tag{3·3}$$

Hence the $|\omega|$-linear contribution of $\langle\langle \tilde{n}_{d\sigma};\ \tilde{n}_{d\sigma'}\rangle\rangle_{i\omega}$ has the form

$$-\frac{|\omega|}{\pi}[\mathrm{Im}\ (G_{ddm\sigma}(+i0)]^2\sum_{mm'}\sum_{m_1\sigma_1}{}'\,\Gamma^{(\sigma\sigma_1\sigma_1)}(m;\ m_1i0,\ m_1i0)$$
$$\times\Gamma^{(\sigma'\sigma_1\sigma_1)}(m';\ m_1i0,\ m_1i0). \tag{3·4}$$

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Here use has been made of the sum $\Gamma^{(\sigma\sigma_1\sigma_1)}$ of all the possible contributions to Γ_1 (or Γ_2).

[3] $\Gamma^{(\sigma\sigma_1\sigma_1)}$ is related to $\partial\Sigma_{dm_1\sigma_1}(i0)/\partial\delta\mu_{d\sigma}|_{\delta\mu=0}$, which appeared in § 2. In fact the following relation holds:

$$\sum_m \Gamma^{(\sigma\sigma_1\sigma_1)}(m;\ m_1 i0,\ m_1 i0) = \delta_{\sigma\sigma_1} - \left.\frac{\partial\,\Sigma_{dm_1\sigma_1}(i0)}{\partial\delta\mu_{d\sigma}}\right|_{\delta\mu=0}. \tag{3·5}$$

(3·5) is a sort of Ward's identity,[9] which connects with a vertex part a change of the proper self-energy part due to an infinitesimally small external perturbation (the spin-dependent impurity chemical potential in the present case). Using (3·4), (3·5) and (2·15) we obtain

$$|\omega|\text{-linear contribution of } \frac{1}{4}\sum_{\sigma\sigma'}\sigma\sigma'\langle\!\langle\tilde{n}_{d\sigma};\ \tilde{n}_{d\sigma'}\rangle\!\rangle_{i\omega}$$

$$= -\frac{|\omega|}{2\pi}[\mathrm{Im}G_{ddm\sigma}(+i0)]^2\sum_m\left(1-\left(\frac{\partial\,\Sigma_{dm\uparrow}}{\partial\delta\mu_{d\uparrow}}-\frac{\partial\,\Sigma_{dm\uparrow}}{\partial\delta\mu_{d\downarrow}}\right)\Bigg|_{\delta\mu=0}\right)^2$$

$$= -\frac{2\pi}{2l+1}|\omega|\,[\langle\!\langle S_d{}^z;\ S_d{}^z\rangle\!\rangle_{i0}]^2$$

$$= \frac{2\pi}{2l+1}i\omega\,(i\cdot\mathrm{sgn}\,\omega)\,[\langle\!\langle S_d{}^z;\ S_d{}^z\rangle\!\rangle_{i0}]^2\,. \tag{3·6}$$

The analytic continuation leads us from (3·6) to

$$\lim_{\omega\to 0}\left[\frac{1}{\omega}\,\mathrm{Im}\,\langle\!\langle S_d{}^z;\ S_d{}^z\rangle\!\rangle_{\omega+i\delta}\right] = \frac{2\pi}{2l+1}[\langle\!\langle S_d{}^z;\ S_d{}^z\rangle\!\rangle_{i0}]^2\,, \tag{3·7}$$

which is exactly the same as (1·4a) and (1·6).

§ 4. The Korringa relation for the *d*-orbital mechanism

The analysis in §§ 2 and 3 can be easily extended to the proof for the *d*-orbital mechanism. We add to H, (1·5), a new term H'_{orb} instead of H'_{spin}, where H'_{orb} is an orbital-dependent perturbation

$$H'_{\mathrm{orb}} = -\sum_{m\sigma}\delta\mu_{dm}d^+_{m\sigma}d_{m\sigma}\,. \tag{4·1}$$

The localized charge Δn_{dm} with the orbital angular momentum m is given by

$$\Delta n_{dm} = \int_{-\infty}^{\infty}d\varepsilon f(\varepsilon)\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_\sigma G_{ddm\sigma}(\varepsilon+i\delta),$$

since the conduction-electron part vanishes for the constant density of states of host conduction electrons. Notice that $G_{ddm\sigma}(\varepsilon+i\delta)$ here is the complete *d*-electron Green's function for $H+H'_{\mathrm{orb}}$. In the same way as in § 2 we transform the above expression into

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$$\Delta n_{dm}=\int_{-\infty}^{\infty}d\varepsilon f(\varepsilon)\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_{\sigma}\left[\frac{\partial}{\partial\varepsilon}\ln(E_{dm}-i\Delta+\Sigma_{dm\sigma}(\varepsilon+i\delta)-\varepsilon)\right.$$

$$\left.+G_{ddm\sigma}(\varepsilon+i\delta)\frac{\partial}{\partial\varepsilon}\Sigma_{dm\sigma}(\varepsilon+i\delta)\right], \tag{4·2}$$

$$E_{dm}\equiv E_d-\delta\mu_{dm}\,.$$

Again we can prove that at $T=0°\mathrm{K}$

$$\int_{-\infty}^{\infty}d\varepsilon f(\varepsilon)\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_{\sigma}\left[G_{ddm\sigma}(\varepsilon+i\delta)\frac{\partial}{\partial\varepsilon}\Sigma_{dm\sigma}(\varepsilon+i\delta)\right]=0 \tag{4·3}$$

holds, because the J-term in (1·5)

$$J\sum_{\substack{mm'\\ \sigma\sigma'}}d^+_{m\sigma}d^+_{m'\sigma'}d_{m\sigma'}d_{m'\sigma}=-J\sum_{\substack{mm'\\ \sigma\sigma'}}d^+_{m\sigma}d^+_{m'\sigma'}d_{m'\sigma}d_{m\sigma'}\,,$$

may be viewed as showing that the spins are exchanged at the interaction vertex, while the orbits are unchanged. Then (4·2) leads to the Friedel sum rule

$$\Delta n_{dm}=\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_{\sigma}\ln(E_{dm}-i\Delta+\Sigma_{dm\sigma}(i\delta))\,. \tag{4·4}$$

From this equation we obtain

$$\left.\frac{\partial\Delta n_{dm}}{\partial\delta\mu_{dm'}}\right|_{\delta\mu=0}=\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_{\sigma}\left[\frac{\delta_{mm'}-(\partial\Sigma_{dm\sigma}(i\delta)/\partial\delta\mu_{dm'})|_{\delta\mu=0}}{-E_d-\Sigma_{dm\sigma}(i\delta)+i\Delta}\right], \tag{4·5}$$

$\Sigma_{dm\sigma}(i\delta)$ in the denominator is independent of m and σ actually. The left-hand side is related to the fluctuation $\langle\langle\tilde{n}_{dm};\tilde{n}_{dm'}\rangle\rangle_{i0}$, where $\tilde{n}_{dm}=\sum_{\sigma}(d^+_{m\sigma}d_{m\sigma}-\langle d^+_{m\sigma}d_{m\sigma}\rangle)$. From (4·5)

$$\langle\langle\tilde{n}_{dm};\tilde{n}_{dm'}\rangle\rangle_{i0}=\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_{\sigma}\left[\frac{\delta_{mm'}-(\partial\Sigma_{dm\sigma}(i\delta)/\partial\delta\mu_{dm'})|_{\delta\mu=0}}{-E_d-\Sigma_{dm\sigma}(i\delta)+i\Delta}\right] \tag{4·6}$$

is obtained. $\partial\Sigma_{dm\sigma}(i\delta)/\partial\delta\mu_{dm'}|_{\delta\mu=0}$ has the following properties: $\partial\Sigma_{dm\sigma}(i\delta)/\partial\delta\mu_{dm}|_{\delta\mu=0}$ is independent of m, and $\partial\Sigma_{dm\sigma}(i\delta)/\partial\delta\mu_{dm'}|_{\delta\mu=0}$ $(m\neq m')$ is also independent of m and m'. This property leads to

$$\langle\langle L^z;L^z\rangle\rangle_{i0}$$

$$=\sum_{mm'}mm'\langle\langle\tilde{n}_{dm};\tilde{n}_{dm'}\rangle\rangle_{i0}$$

$$=\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\sum_{\sigma}\left(\sum_{m_1}m_1^2\right)\left[\frac{1-((\partial\Sigma_{dm\sigma}(i\delta)/\partial\delta\mu_{dm})-(\partial\Sigma_{dm\sigma}(i\delta)/\partial\delta\mu_{dm'}))|_{\delta\mu=0}}{-E_d-\Sigma_{dm\sigma}(i\delta)+i\Delta}\right].$$

$$(m'\neq m) \tag{4·7}$$

Now we turn to the $|\omega|$-linear contribution of $\langle\langle L^z;L^z\rangle\rangle_{i\omega}$ at small ω. The equation corresponding to (3·4) is given by

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$|\omega|$-linear part of $\langle\!\langle \tilde{n}_{dm};\ \tilde{n}_{dm'}\rangle\!\rangle_{i\omega}$

$$= -\frac{|\omega|}{\pi}[\mathrm{Im}\ G_{ddm\sigma}(i\delta)]^2 \sum_{\sigma\sigma'}\sum_{m_1\sigma_1} \Gamma^{(\sigma\sigma_1\sigma_1)}(m;\ m_1 i0,\ m_1 i0)$$
$$\times \Gamma^{(\sigma'\sigma_1\sigma_1)}(m';\ m_1 i0,\ m_1 i0) \qquad (4\cdot 8)$$

and instead of $(3\cdot 6)$ we have

$$\sum_{\sigma} \Gamma^{(\sigma\sigma_1\sigma_1)}(m;\ m_1 i0,\ m_1 i0) = \delta_{mm_1} - \frac{\partial \Sigma_{dm_1\sigma_1}}{\partial \delta\mu_{dm}}\bigg|_{\delta\mu=0}. \qquad (4\cdot 9)$$

Combining $(4\cdot 8)$ with $(4\cdot 9)$,

$|\omega|$-linear contribution of $\langle\!\langle L^z;\ L^z\rangle\!\rangle_{i\omega}$

$$= -\frac{2|\omega|}{\pi}[\mathrm{Im}\ G_{ddm\sigma}(i\delta)]^2 \left(\sum_{m_1} m_1^2\right)\left[1-\left(\frac{\partial \Sigma_{dm\sigma}}{\partial\delta\mu_{dm}} - \frac{\partial \Sigma_{dm\sigma}}{\partial\delta\mu_{dm'}}\right)\bigg|_{\delta\mu=0}\right]^2$$
$$= -2\pi|\omega|\frac{1}{(4/3)\,l(l+1)\,(2l+1)}[\langle\!\langle L^z;\ L^z\rangle\!\rangle_{i0}]^2$$

can be obtained. One may write it in the form

$$\lim_{\omega\to 0}\left[\frac{1}{\omega}(\mathrm{Im}\ \langle\!\langle L^z;\ L^z\rangle\!\rangle_{\omega+i\delta})\right] = \frac{2\pi}{(4/3)\,l(l+1)\,(2l+1)}[\langle\!\langle L^z;\ L^z\rangle\!\rangle_{i0}]^2, \qquad (4\cdot 10)$$

which is the KR for the d-orbital mechanism.

§ 5. Supplementary discussion

(1) We have proved the Korringa relation (KR) for the impurity nuclear spin relaxation due to the d-spin mechanism as well as the d-orbital mechanism. Our proof has shown that for the model $(1\cdot 5)$ the KR is valid at low temperatures in any order of the perturbation expansion. It also suggests that the KR may hold for a Hamiltonian more general than $(1\cdot 5)$. If the exchange enhancement is present in the host, however, the KR breaks down even in the RPA.[10] Therefore the locality of the Coulomb and exchange interaction such as in $(1\cdot 5)$ is certainly one of the necessary conditions for the KR.*) In this connection we want to make a comment on the assumption of the constant density of states of conduction electrons. Actually that assumption is not essential to our proof, although it is quite reasonable for transition-metal impurities in simple metals. If we do not use the assumption, we have to take into account the conduction-electron part $\sum_k (n_{k\sigma} - n_{k\sigma}^0)$ of the localized charge; $(2\cdot 13)$ should be then replaced by

*) The author is indebted to Professor K. Yosida and Professor A. Yoshimori for calling his attention to this point.

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$$\left.\frac{\partial \Delta n_{d\sigma}}{\partial \delta \mu_{d\sigma'}}\right|_{\delta\mu=0} = \langle\langle \tilde{n}_{d\sigma} + \tilde{n}_{c\sigma};\ \tilde{n}_{d\sigma'} \rangle\rangle\,, \tag{5·1}$$

where $\tilde{n}_{c\sigma} = \sum_k (a_{k\sigma}^+ a_{k\sigma} - \langle a_{k\sigma}^+ a_{k\sigma}\rangle)$. Therefore $\langle\langle S_d^z;\ S_d^z\rangle\rangle_{i0}$ in (3·6) and (3·7) is taken over by $\langle\langle S_d^z;\ S_d^z + S_c^z\rangle\rangle_{i0}$, where $S_c^z \equiv (1/2)\sum_{k\sigma}\sigma a_{k\sigma}^+ a_{k\sigma}$. Instead of (3·7) we obtain

$$\lim_{\omega\to 0}\left[\frac{1}{\omega}\,\mathrm{Im}\,\langle\langle S_d^z;\ S_d^z\rangle\rangle_{\omega+i\delta}\right] = \frac{2\pi}{2l+1}[\langle\langle S_d^z;\ S_d^z + S_c^z\rangle\rangle_{i0}]^2\,. \tag{5·2}$$

$\langle\langle S_d^z;\ S_d^z + S_c^z\rangle\rangle_{i0}$ is a d-spin induced by a small uniform magnetic field, and it is exactly the quantity that appears in the Knight shift $K_{(d)}$. It is also easy to prove the KR for the Wolff model.[11] The outline of the proof is given in the Appendix. Anyhow the proof of the KR leads us to conclude that the KR must be valid in a wide variety of systems, as far as the host exchange is negligible and the conditions, $T \ll T_K$ and $H \ll H_K$, are satisfied. We refer the reader to Refs. 4) and 5) as to the analysis of experimental data based on the KR.

(2) In § 3 we have examined the low-frequency dynamical spin susceptibility and showed that it is related to the static spin susceptibility. It is easy to see that an analogous relation holds for the *charge fluctuations*:

$$\lim_{\omega\to 0}\mathrm{Im}\left[\frac{\langle\langle \frac{1}{2}(\tilde{n}_{d\uparrow}+\tilde{n}_{d\downarrow});\ \frac{1}{2}(\tilde{n}_{d\uparrow}+\tilde{n}_{d\downarrow})\rangle\rangle_{\omega+i\delta}}{\omega}\right]$$
$$= \frac{2\pi}{2l+1}\left[\left\langle\!\!\left\langle \frac{1}{2}(\tilde{n}_{d\uparrow}+\tilde{n}_{d\downarrow});\ \frac{1}{2}(\tilde{n}_{d\uparrow}+\tilde{n}_{d\downarrow})\right\rangle\!\!\right\rangle_{i0}\right]^2. \tag{5·3}$$

The static charge susceptibility is suppressed by the Coulomb repulsion; consequently the low-frequency charge fluctuation is also suppressed as in (5·3).

(3) The static spin susceptibility is enhanced by the Coulomb and exchange interaction. The Korringa relation shows that the low-frequency spin fluctuations are also enhanced. Using the perturbation expansion, Yamada estimated the enhancement of the static susceptibility for the symmetric single-band model. The corresponding low-frequency spin fluctuations are easily evaluated with the aid of (3·7). It is expected in general that $\langle\langle S_d^z;\ S_d^z\rangle\rangle_{i0} \propto T_{K\,\mathrm{spin}}^{-1}$. It leads to

$$\lim_{\omega\to 0}\mathrm{Im}[(\chi_{\mathrm{spin}}(\omega+i\delta)/\omega] \propto T_{K\,\mathrm{spin}}^{-2}\,.$$

(4) In this paper we have studied only the low-frequency spin and orbital susceptibilities at zero temperature in order to prove the KR. The spin (orbital moment) at zero temperature is quenched at low frequencies, i.e., $\omega \ll T_{K\,\mathrm{spin}}$ ($\omega \ll T_{K\,\mathrm{orb}}$). However if we look at the impurity with a time-scale shorter than $\hbar/k_B T_K$ an appreciable spin or orbital moment should be expected. In order to confirm this picture we are currently studying the frequency dependence of $\chi(\omega+i\delta)$, the spectrum Im $\chi(\omega+i\delta)$ in particular, over the whole ω region. It will be published in a forthcoming paper.

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Appendix

——The Korringa relation in the Wolff model——

Here we want to give the outline of the proof of the Korringa relation (KR) for the single-band Wolff model:[11]

$$H=\sum_{ij} t_{ij}a_{i\sigma}^{+}a_{j\sigma}+V\sum_{\sigma} a_{0\sigma}^{+}a_{0\sigma}+Ua_{0\uparrow}^{+}a_{0\downarrow}^{+}a_{0\downarrow}a_{0\uparrow}\,, \tag{A·1}$$

where $a_{i\sigma}$ is the annihilation operator of an electron at the j-th site. The transfer matrix t_{ij} is arbitrary, and the impurity is assumed to be located at the origin. The KR for this model reads[12]

$$\lim_{\omega\to 0}\left[\frac{1}{\omega}\,\mathrm{Im}\,\langle\langle S_0^z;\ \dot{S}_0^z\rangle\rangle_{\omega+i\delta}\right]=2\pi[\langle\langle S_0^z;\ \sum_j S_j^z\rangle\rangle_{i0}]^2\,, \tag{A·2}$$

where S_j^z is the spin operator at the j-th site, i.e., $(a_{j\uparrow}{}^{+}a_{j\uparrow}-a_{j\downarrow}{}^{+}a_{j\downarrow})/2$. The KR (A·2), which corresponds to (5·2) in the Anderson model, was proved by Lederer and Mills[12] by using the RPA. However, as we show below, the relation is quite general.

By adding a small perturbation $H'_{\mathrm{spin}}=-\sum_\sigma \delta\mu_{0\sigma}a_{0\sigma}^{+}a_{0\sigma}$ to (A·1) we study the total displaced charge Δn_σ for the spin σ. Δn_σ can be written as

$$\Delta n_\sigma=\int_{-\infty}^{\infty}d\varepsilon f(\varepsilon)\left(-\frac{1}{\pi}\,\mathrm{Im}\right)[\sum_k (G_k{}^0(\varepsilon+i\delta))^2 t_\sigma(\varepsilon+i\delta)], \tag{A·3}$$

where $G_k{}^0(\varepsilon+i\delta)=[\varepsilon+i\delta-\varepsilon_k]^{-1}$ (ε_k=Fourier transform of t_{ij}) is the host Green's function. $t_\sigma(\varepsilon+i\delta)$ is the t-matrix of the impurity scattering, i.e.,

$$t_\sigma(\varepsilon+i\delta)=\frac{V_\sigma}{1-V_\sigma G_{00}^0(\varepsilon+i\delta)}$$
$$+\frac{1}{(1-V_\sigma G_{00}^0(\varepsilon+i\delta))^2}\,\frac{\Sigma_\sigma(\varepsilon+i\delta)}{1-\Sigma_\sigma(\varepsilon+i\delta)G_{00}^0(\varepsilon+i\delta)/(1-V_\sigma G_{00}^0(\varepsilon+i\delta))}\,, \tag{A·4}$$

where $V_\sigma\equiv V-\delta\mu_{0\sigma}$ and $G_{00}^0(\varepsilon+i\delta)=\sum_k G_k{}^0(\varepsilon+i\delta)$. The first term of (A·4) is a pure potential-scattering, while the second term describes the many-body scattering modified by the potential scattering. The self-energy $\Sigma_\sigma(\varepsilon+i\delta)$ is defined as

$$\overline{G}_{00\sigma}(\varepsilon+i\delta)=\frac{G_{00}^0(\varepsilon+i\delta)}{1-(V_\sigma+\Sigma_\sigma(\varepsilon+i\delta))G_{00}^0(\varepsilon+i\delta)} \tag{A·5}$$

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with the propagator $\overline{G}_{00\sigma}$ from the origin to the origin in the alloy. Substituting (A·4) into (A·3) and using the Luttinger's relation at $T=0°$K

$$\int_{-\infty}^{\infty} d\varepsilon f(\varepsilon)\left(-\frac{1}{\pi}\,\mathrm{Im}\right)\left[\overline{G}_{00\sigma}(\varepsilon+i\delta)\frac{\partial\Sigma_\sigma(\varepsilon+i\delta)}{\partial\varepsilon}\right]=0 \qquad (A\cdot 6)$$

for the present case, we find that at $T=0°$K

$$\Delta n_\sigma=\left(-\frac{1}{\pi}\right)\mathrm{Im}\,\{\ln[1-(V_\sigma+\Sigma_\sigma(i0))\,G_{00}^0(i0)]\}$$

$$=\frac{1}{\pi}\delta_\sigma. \qquad (A\cdot 7)$$

Here the phase shift at the Fermi energy

$$\delta_\sigma=\tan^{-1}\frac{(V_\sigma+\Sigma_\sigma(i0))\,I_{00}}{(V_\sigma+\Sigma_\sigma(i0))\,R_{00}-1} \qquad (A\cdot 8)$$

was introduced with $G_{00}^0(i0)=R_{00}-iI_{00}$. (A·7) is the Friedel sum rule in the present case. From (A·7) we obtain

$$\left.\frac{\partial\Delta n_\sigma}{\partial\delta\mu_{0\sigma'}}\right|_{\delta\mu=0}=\left(-\frac{1}{\pi}\right)\mathrm{Im}[\overline{G}_{00\sigma}(i0)]\left(\delta_{\sigma\sigma'}-\left.\frac{\partial\Sigma_\sigma(i0)}{\partial\delta\mu_{0\sigma'}}\right|_{\delta\mu=0}\right). \qquad (A\cdot 9)$$

The left-hand side is equivalent to $\langle\langle\tilde{n}_{0\sigma'};\ \sum_j\tilde{n}_{j\sigma}\rangle\rangle_{i0}$, where $\tilde{n}_{j\sigma}\equiv a_{j\sigma}^\dagger a_{j\sigma}-\langle a_{j\sigma}^\dagger a_{j\sigma}\rangle$.

The analysis of the low-frequency thermodynamic Green's function $\langle\langle n_{0\sigma};\ n_{0\sigma'}\rangle\rangle_{i\omega}$ can be made in the same way as in § 3. The only thing we have to do is to omit the index m and to replace $G_{d\sigma}^0(i\varepsilon)$ by $G_{00}^0(i\varepsilon)/(1-VG_{00}^0(i\varepsilon))$. Therefore we easily find

$$|\omega|\text{-linear part of } \langle\langle n_{0\sigma};\ n_{0\sigma'}\rangle\rangle_{i\omega}$$

$$=-\frac{|\omega|}{\pi}[\mathrm{Im}[\overline{G}_{00}(i0)]]^2\sum_{\sigma_1}\left(\delta_{\sigma\sigma_1}-\left.\frac{\partial\Sigma_{\sigma_1}(i0)}{\partial\delta\mu_{0\sigma}}\right|_{\delta\mu=0}\right)$$

$$\times\left(\delta_{\sigma'\sigma_1}-\left.\frac{\partial\Sigma_{\sigma_1}(i0)}{\partial\delta\mu_{0\sigma'}}\right|_{\delta\mu=0}\right). \qquad (A\cdot 10)$$

Combining (A·10) with (A·9) we obtain (A·2).

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