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Dynamical Theory of Spin Waves in Ferromagnetic Metals with *s-d* Exchange Interaction

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The spectrum of spin waves in ferromagnetic metals where ferromagnetism is caused by the so-called *s-d* exchange interaction between localized spins and conduction electrons is investigated theoretically without taking an adiabatic approximation which is usually used. The dynamical susceptibility of the system is calculated by making use of the two-time Green's function, and the spectrum is determined as divergent points of the susceptibility. As non-adiabatic effects, there appear some features in the spectrum which have not been considered in usual adiabatic theories. First, if the wave number of spin waves is not so large, there are two or more modes of collective oscillations of the system, among which one may well be called acoustical mode where two spin systems, localized spins and conduction electron spins, oscillate in phase, and another an optical mode where they oscillate in antiphase. Further, as to the acoustical mode, the spectrum has a dip in the vicinity of a certain wave number where an adiabatic approximation becomes incorrect.

§ 1. Introduction

Magnetic properties of the system where localized spins and conduction electrons interact with each other via the so-called *s-d* exchange interaction have been investigated theoretically by several authors.[1),2),3),4)] It is found that there appears an effective interaction between localized spins which is obtained by the second order perturbation. To get the effective interaction, most of the authors[1),2),3)] took an adiabatic approximation or the Born-Oppenheimer approximation; i.e. the motion of conduction electrons was assumed to be much faster than that of localized spins and the latter was ignored compared with the former. As pointed out by Hasegawa,[5)] however, the approximation can not always be allowed, since, unlike the case of the interaction between nuclei in molecules, the energy spectrum of conduction electrons is continuous and the energy difference of conduction electrons may be smaller than the frequency of the motion of localized spins. Therefore the problem is worth discussing more carefully.

We shall consider, in this paper, a ferromagnetic metal where ferromagnetism is caused by the above-mentioned interaction and investigate theoretically the spectrum of spin waves in the system. The problem was already discussed by Kasuya[2)] in the adiabatic approximation. Our object is now to see how non-adiabatic effects appear in the spectrum.

In § 2 we shall calculate the dynamical susceptibility of the system by the use of two-time Green's function.[6] The spectrum of spin waves is determined as divergent points of the susceptibility, which will be given in § 3. In our treatment, spin waves have a finite lifetime and we shall calculate it in § 4. Finally, in § 5, some arguments and additional remarks will be given briefly.

§ 2. Dynamical susceptibility

Let us consider a system consisting of N spins, localized regularly at lattice sites, and N band electrons. The so-called *s-d* exchange interaction between localized spins and band electrons are taken into account, but direct interactions between localized spins and between band electrons are ignored. Then the Hamiltonian of this system is given as follows:

$$H=\sum_{nk}\sum_{\sigma}\epsilon_{nk}\, a_{nk\sigma}^{*}\, a_{nk\sigma}-N^{-1}\sum_{nn'}\sum_{kk'} J_{nn'}(\boldsymbol{k},\boldsymbol{k}')\,[(a_{n'k'+}^{*}\, a_{nk+}-a_{n'k'-}^{*}\, a_{nk-})\, S^{z}(\boldsymbol{k}'-\boldsymbol{k})$$
$$+a_{n'k'+}^{*}\, a_{nk-}\, S^{-}(\boldsymbol{k}'-\boldsymbol{k})+a_{n'k'-}^{*}\, a_{nk+}\, S^{+}(\boldsymbol{k}'-\boldsymbol{k})], \tag{1}$$

where

$$S^{\alpha}(\boldsymbol{k})=\sum_{\lambda} S_{\lambda}^{\alpha}\exp(-i\boldsymbol{k}\cdot\boldsymbol{R}_{\lambda}).$$

Here $a_{nk\sigma}^{*}$ and $a_{nk\sigma}$ are, respectively, the ordinary creation and annihilation operators of the n-th band electron with wave vector $\boldsymbol{k}$ and spin σ, and ϵ_{nk} is its one electron energy measured from chemical potential μ, S_{λ}^{α} is the α component of the localized spin at $\boldsymbol{R}_{\lambda}$ site, and $J_{nn'}(\boldsymbol{k},\boldsymbol{k}')$ is the matrix element of the *s-d* exchange interaction. This Hamiltonian is an obvious generalization of Kasuya's one:[2] i.e. interband terms are contained explicitly in our Hamiltonian.

In order to investigate magnetic properties of the system, we have only to calculate the dynamical susceptibility $\chi(\boldsymbol{q},\omega)$, which is given by[7]

$$\chi^{+-}(\boldsymbol{q},\omega)=i\int_{0}^{\infty}dt\, e^{i\omega t}\langle[M^{+}(\boldsymbol{q},t),\, M^{-}(-\boldsymbol{q})]\rangle, \tag{2}$$

where

$$M^{\pm}(\boldsymbol{q})=2\mu_{B}\Sigma^{\pm}(\boldsymbol{q})$$
$$=2\mu_{B}\{S^{\pm}(\boldsymbol{q})+\sum_{nn'}\sum_{k} a_{n'k-q\pm}^{*}\, a_{nk\mp}\}, \tag{3}$$

(μ_B: Bohr magneton)

$$\langle A\rangle=\mathrm{Tr}(e^{-\beta H}A)/\mathrm{Tr}(e^{-\beta H}),\ \text{and}\ A(t)=e^{iHt}Ae^{-iHt}.$$

Here we have taken $\hbar=1$. In Eq. (3) the summation $\sum_{k}$ runs over the first Brillouin zone and, if $\boldsymbol{k}-\boldsymbol{q}$ is not in the first zone, it must be reduced to the first zone. Precisely speaking, in the expression (2), we must replace ω by $\omega+i\varepsilon$ and take the limit $\varepsilon\to+0$ after calculation. The same abbreviation as in

(2) will be used in the following.

Now we introduce two-time Green's function defined by[6)]

$$\langle\langle A(t);B\rangle\rangle=\begin{cases}-i\langle[A(t),\ B]\rangle & \text{for } t>0,\\ 0 & \text{for } t<0,\end{cases} \tag{4}$$

and its Fourier transform

$$\langle\langle A;B\rangle\rangle_\omega=\int_{-\infty}^{\infty}dt\, e^{i\omega t}\langle\langle A(t);B\rangle\rangle. \tag{5}$$

Then Eq. (2) can be rewritten as

$$\begin{aligned}\chi^{+-}(\boldsymbol{q},\omega)&=-(2\mu_B)^2\langle\langle\Sigma^+(\boldsymbol{q});\ \Sigma^-(-\boldsymbol{q})\rangle\rangle_\omega\\ &=-(2\mu_B)^2\{\langle\langle S^+(\boldsymbol{q})\rangle\rangle_\omega+\sum_{nn'}\sum_{k}\langle\langle a^*_{n'k-q+}\,a_{nk-}\rangle\rangle_\omega\},\end{aligned} \tag{6}$$

where the abbreviation

$$\langle\langle A;\Sigma^-(-\boldsymbol{q})\rangle\rangle_\omega\equiv\langle\langle A\rangle\rangle_\omega$$

has been used. The problem is now reduced to the calculation of Green's functions $\langle\langle S^+(\boldsymbol{q})\rangle\rangle_\omega$ and $\langle\langle a^*_{n'k'+}a_{nk-}\rangle\rangle_\omega$.

It is easy to find the equations of motion obeyed by these Green's functions, which are given from the Hamiltonian (1) as follows:

$$\begin{aligned}\omega\langle\langle S^+(\boldsymbol{q})\rangle\rangle_\omega=2NS-N^{-1}\sum_{nn'}\sum_{kk'}J_{nn'}(\boldsymbol{k},\boldsymbol{k}')\,\{&2\langle\langle a^*_{n'k'+}\,a_{nk-}\,S^z(\boldsymbol{k}'-\boldsymbol{k}+\boldsymbol{q})\rangle\rangle_\omega\\ &-\langle\langle(a^*_{n'k'+}\,a_{nk+}-a^*_{n'k'-}\,a_{nk-})S^+(\boldsymbol{k}'-\boldsymbol{k}+\boldsymbol{q})\rangle\rangle_\omega\},\end{aligned} \tag{7}$$

$$\begin{aligned}\omega\langle\langle a^*_{n'k-q+}a_{nk-}\rangle\rangle=&\sum_m[\langle a^*_{n'k-q+}\,a_{mk-q+}\rangle-\langle a^*_{mk-}\,a_{nk-}\rangle]\\ &+[\epsilon_{nk}-\epsilon_{n'k-q}]\langle\langle a^*_{n'k-q+}\,a_{nk-}\rangle\rangle_\omega\\ &-N^{-1}\sum_{mp}\{J_{mn}(\boldsymbol{p},\boldsymbol{k})\,[\langle\langle a^*_{n'k-q+}\,a_{mp+}S^+(\boldsymbol{k}-\boldsymbol{p})\rangle\rangle_\omega-\langle\langle a^*_{n'k-q+}\,a_{mp-}S^z(\boldsymbol{k}-\boldsymbol{p})\rangle\rangle_\omega]\\ &-J_{n'm}(\boldsymbol{k}-\boldsymbol{q},\boldsymbol{p})\,[\langle\langle a^*_{mp-}a_{nk-}S^+(\boldsymbol{p}-\boldsymbol{k}+\boldsymbol{q})\rangle\rangle_\omega+\langle\langle a^*_{mp+}\,a_{nk-}\,S^z(\boldsymbol{p}-\boldsymbol{k}+\boldsymbol{q})\rangle\rangle_\omega]\}.\end{aligned} \tag{8}$$

Here it has been assumed that the system is ferromagnetic at low temperatures and that the temperature is low enough for us to take

$$\langle S_\lambda^z\rangle=S, \tag{9}$$

where S is the magnitude of the localized spin.

To solve Eqs. (7) and (8), and to determine Green's functions $\langle\langle S^+(\boldsymbol{q})\rangle\rangle_\omega$ and $\langle\langle a^*_{n'k-q+}a_{nk-}\rangle\rangle_\omega$, it is required to take some approximations for the higher order Green's functions which appear on the right-hand sides of Eqs. (7) and (8). We put

$$\left.\begin{aligned}&\langle\langle a^*_{n'k'+}\,a_{nk-}S^z(\boldsymbol{k}'-\boldsymbol{k}+\boldsymbol{q})\rangle\rangle_\omega=\delta_{k',k-q}\langle S^z(0)\rangle\langle\langle a^*_{n'k-q+}\,a_{nk-}\rangle\rangle_\omega,\\ &\langle\langle a^*_{n'k'\sigma}\,a_{nk\sigma}\,S^+(\boldsymbol{k}'-\boldsymbol{k}+\boldsymbol{q})\rangle\rangle_\omega=\delta_{k,k'}\langle a^*_{n'k\sigma}\,a_{nk\sigma}\rangle\langle\langle S^+(\boldsymbol{q})\rangle\rangle_\omega,\end{aligned}\right\} \tag{10}$$

and so on, which is considered as a kind of the random phase approximation. It should be noticed here that $\langle a_{n'k\sigma}^{*} a_{nk\sigma}\rangle$ dose not vanish even for $n'\neq n$ because of the interband terms of the *s-d* exchange interaction.

If Green's function

$$\langle\!\langle a_{nk\sigma}\,;\ a_{n'k'\sigma}^{*}\rangle\!\rangle_{\omega}=\frac{1}{i}\int_{0}^{\infty} dt\, e^{i\omega t}\langle[a_{nk\sigma}(t),\ a_{n'k\sigma}^{*}]_{+}\rangle \tag{11}$$

is known, the average $\langle a_{n'k\sigma}^{*} a_{nk\sigma}\rangle$ can be obtained by the use of formula[6)]

$$\langle a_{n'k\sigma}^{*}\, a_{nk\sigma}\rangle=i\int_{-\infty}^{\infty}\frac{d\omega}{e^{\beta\omega}+1}\{\langle\!\langle a_{nk\sigma}\,;\ a_{n'k\sigma}^{*}\rangle\!\rangle_{\omega+i\varepsilon}-\langle\!\langle a_{nk\sigma}\,;\ a_{n'k\sigma}^{*}\rangle\!\rangle_{\omega-i\varepsilon}\}. \tag{12}$$

$$(\varepsilon\to+0.)$$

Making use of the Hamiltonian (1) and the same approximation as (10), we get the equation for Green's function (11) as follows:

$$\omega\langle\!\langle a_{nk\pm}\,;\ a_{n'k\pm}^{*}\rangle\!\rangle_{\omega}=\frac{1}{2\pi}\delta_{nn'}+\epsilon_{nk}\langle\!\langle a_{nk\pm}\,;\ a_{n'k\pm}^{*}\rangle\!\rangle_{\omega}$$
$$\mp S\sum_{m}J_{mn}(\boldsymbol{k},\boldsymbol{k})\langle\!\langle a_{mk\pm}\,;\ a_{n'k\pm}^{*}\rangle\!\rangle_{\omega}, \tag{13}$$

which is solved to the first order of $J_{n'n}(n'\neq n)$ as

$$\left.\begin{aligned}&\langle\!\langle a_{nk\sigma}\,;\ a_{nk\sigma}^{*}\rangle\!\rangle_{\omega}=\frac{1}{2\pi}\,\frac{1}{\omega-\epsilon_{nk\sigma}},\\&\langle\!\langle a_{nk\pm}\,;\ a_{n'k\pm}^{*}\rangle\!\rangle_{\omega}=\mp\frac{1}{2\pi}\,\frac{J_{n'n}(\boldsymbol{k},\boldsymbol{k})}{(\omega-\epsilon_{nk\pm})(\omega-\epsilon_{n'k\pm})},\ \text{for}\ n\neq n',\end{aligned}\right\} \tag{14}$$

with

$$\epsilon_{nk\pm}=\epsilon_{nk}\mp J_{nn}(\boldsymbol{k},\boldsymbol{k})S. \tag{15}$$

Therefore

$$\left.\begin{aligned}&\langle a_{nk\sigma}^{*}\, a_{nk\sigma}\rangle=f_{nk\sigma},\\&\langle a_{n'k\pm}^{*}\, a_{nk\pm}\rangle=\mp J_{n'n}(\boldsymbol{k},\boldsymbol{k})S\frac{f_{nk\pm}-f_{n'k\pm}}{\epsilon_{nk\pm}-\epsilon_{n'k\pm}},\ \text{for}\ n\neq n',\end{aligned}\right\} \tag{16}$$

where $f_{nk\sigma}=f(\epsilon_{nk\sigma})$ and $f(\epsilon)$ is the Fermi distribution function.

Inserting Eqs. (10) and (16) into Eqs. (7) and (8), we get the simultaneous equations for Green's functions $\langle\!\langle S^{+}(\boldsymbol{q})\rangle\!\rangle$ and $\langle\!\langle a_{n'k-q+}^{*}a_{nk-}\rangle\!\rangle$ as follows:

$$\omega\langle\!\langle S^{+}(\boldsymbol{q})\rangle\!\rangle_{\omega}=2NS-2S\sum_{nn'}\sum_{k}J_{nn'}(\boldsymbol{k},\boldsymbol{k}-\boldsymbol{q})\langle\!\langle a_{n'k-q+}^{*}\, a_{nk-}\rangle\!\rangle_{\omega}$$
$$+\frac{1}{N}\Big\{\sum_{n}\sum_{k}J_{nn}(\boldsymbol{k},\boldsymbol{k})(f_{nk+}-f_{nk-})-\sum_{n\neq n'}\sum_{k}|J_{n'n}(\boldsymbol{k},\boldsymbol{k})|^{2}S$$
$$\times\left(\frac{f_{nk+}-f_{n'k+}}{\epsilon_{nk+}-\epsilon_{n'k+}}+\frac{f_{nk-}-f_{n'k-}}{\epsilon_{nk-}-\epsilon_{n'k-}}\right)\Big\}\langle\!\langle S^{+}(\boldsymbol{q})\rangle\!\rangle_{\omega}, \tag{17}$$

$$\omega\langle\!\langle a_{n'k-q+}^{*}a_{nk-}\rangle\!\rangle_{\omega}=(f_{n'k-q+}-f_{nk-})+(\epsilon_{n'k-q+}-\epsilon_{nk-})\langle\!\langle a_{n'k-q+}^{*}\ a_{nk-}\rangle\!\rangle_{\omega}$$

$$-\frac{1}{N}J_{n'n}(\boldsymbol{k}-\boldsymbol{q},\boldsymbol{k})(f_{n'k-q+}-f_{nk-})\langle\langle S^{+}(\boldsymbol{q})\rangle\rangle_{\omega}. \tag{18}$$

In the calculation of Eq. (18) we have ignored interband contributions, which is permitted in the lowest approximation. Suppose that the conduction band is not degenerate, and that ω and $J_{n'n}$ are small compared with the interband energy difference $\epsilon_{n'}-\epsilon_{n}$. Then we can put

$$(\epsilon_{n'k-q+}-\epsilon_{nk-})+\omega\cong\epsilon_{n'k-q}-\epsilon_{nk},\ \text{for}\ n\neq n'. \tag{19}$$

The simultaneous equations (17) and (18) can be solved easily, and, using the above approximation and further neglecting small interband contributions which appear in the numerator, the final expression for the dynamical susceptibility $\chi(\boldsymbol{q},\omega)$ is found from Eq. (6) to be

$$\chi^{+-}(\boldsymbol{q},\omega)=-(2\mu_B)^2\frac{N}{D(\boldsymbol{q},\omega)}\{2S+\sum_{nk}[\omega-\frac{1}{N}\sum_{n'k'}J_{n'n'}(\boldsymbol{k}',\boldsymbol{k}')(f_{n'k'+}-f_{n'k'-})$$
$$-2S(J_{nn}(\boldsymbol{k},\boldsymbol{k}-\boldsymbol{q})+J_{nn}(\boldsymbol{k}-\boldsymbol{q},\boldsymbol{k}))]\frac{f_{nk-q+}-f_{nk-}}{\epsilon_{nk-q+}-\epsilon_{nk-}+\omega}\}, \tag{20}$$

where

$$D(\boldsymbol{q},\omega)=\omega-\frac{1}{N}\sum_{nk}J_{nn}(\boldsymbol{k},\boldsymbol{k})(f_{nk+}-f_{nk-})+\frac{2S}{N}\{\sum_{n\neq n'}\sum_{k}|J_{nn'}(\boldsymbol{k},\boldsymbol{k})|^2\frac{f_{nk}-f_{n'k}}{\epsilon_{nk}-\epsilon_{n'k}}$$
$$-\sum_{nk}|J_{nn}(\boldsymbol{k},\boldsymbol{k}-\boldsymbol{q})|^2\frac{f_{nk-q+}-f_{nk-}}{\epsilon_{nk-q+}-\epsilon_{nk-}+\omega}-\sum_{n\neq n'}\sum_{k}|J_{nn'}(\boldsymbol{k},\boldsymbol{k}-\boldsymbol{q})|^2\frac{f_{n'k-q}-f_{nk}}{\epsilon_{n'k-q}-\epsilon_{nk}}\}. \tag{21}$$

Our result as well as our method is essentially the same as Potakov and Tyablikov's one,[4] except that we have taken into account interband terms.

§ 3. Spectrum of spin waves

The spectrum of spin waves is given by the divergent point of the dynamical susceptibility in the preceding section, that is by the relation

$$D(\boldsymbol{q},\omega)=0,$$

which determines the frequency ω as a function of the wave vector $\boldsymbol{q}$. This equation is rewritten as

$$\omega-\omega_{ad}(\boldsymbol{q})=F(\boldsymbol{q},\omega), \tag{22}$$

where

$$\omega_{ad}(\boldsymbol{q})=\frac{1}{N}\{\sum_{nk}J_{nn}(\boldsymbol{k},\boldsymbol{k})(f_{nk+}-f_{nk-})-2S|J_{nn}(\boldsymbol{k},\boldsymbol{k}-\boldsymbol{q})|^2\frac{f_{nk-q+}-f_{nk-}}{\epsilon_{nk-q+}-\epsilon_{nk-}}\}$$
$$+\frac{2S}{N}\sum_{n\neq n'}\sum_{k}\{|J_{nn'}(\boldsymbol{k},\boldsymbol{k})|^2\frac{f_{nk}-f_{n'k}}{\epsilon_{nk}-\epsilon_{n'k}}-|J_{nn'}(\boldsymbol{k},\boldsymbol{k}-\boldsymbol{q})|^2\frac{f_{n'k-q}-f_{nk}}{\epsilon_{n'k-q}-\epsilon_{nk}}\}, \tag{23}$$

and

$$F(\boldsymbol{q},\omega)=\frac{2S}{N}\sum_{nk}|J_{nn}(\boldsymbol{k},\boldsymbol{k}-\boldsymbol{q})|^2\left\{\frac{f_{nk-q+}-f_{nk-}}{\epsilon_{nk-q+}-\epsilon_{nk-}+\omega}-\frac{f_{nk-q+}-f_{nk-}}{\epsilon_{nk-q+}-\epsilon_{nk-}}\right\}. \tag{24}$$

Here $\omega_{ad}(\boldsymbol{q})$ is the adiabatic spectrum of spin waves, while there arise non-adiabatic effects from $F(\boldsymbol{q},\omega)$. If we ignore ω in the function $F(\boldsymbol{q},\omega)$, in other words, if we take the adiabatic approximation, $\omega=\omega_{ad}(\boldsymbol{q})$ is the solution of Eq. (22) since $F(\boldsymbol{q},0)=0$.

The expression (23) of $\omega_{ad}(\boldsymbol{q})$ is equal to the spectrum obtained by Kasuya (cf. Eq. (5·5) of reference 2)), except that we considered the band structure of conduction electrons explicitly. On the other hand, he assumed the spectrum ϵ_{nk} to be free electron-like in an extended zone scheme. Then the summation over bands was replaced there by the summation over reciprocal lattice vectors, though Umklapp processes are included automatically in the latter expression.

Recently Woll and Nettel[8] calculated the spin wave spectra of rare earth metals using Kasuya's expression, and showed that the frequency is negative in some directions of the wave vector. Though a quantitative meaning can hardly be attributed to their calculation because of the assumption on the spectrum of band electrons which can hardly be expected in real metals, we can see easily from the expression (23) that interband contributions to $\omega_{ad}(\boldsymbol{q})$ are negative if the energy maximum (not minimum) is at $\boldsymbol{k}=0$ in excited bands. Therefore, ω_{ad} may be negative if the spectrum ϵ_{nk} is appropriate. Even in such a case, however, if there are appropriate anisotropy energies, ω_{ad} becomes positive again and the ferromagnetic state is stable at low temperatures. We remark here that, even in the presence of such anisotropy energies and moreover of a direct exchange interaction between localized spins, we need not modify Eq. (22) except for the functional form of $\omega_{ad}(\boldsymbol{q})$. We do not discuss on this point further and do not assume the concrete functional form of $\omega_{ad}(\boldsymbol{q})$, but only assume it to be positive for all values of q.

Our problem is now to see how non-adiabatic effects appear in the spectrum of spin waves. For simplicity, it is assumed that f_{nk} does not vanish only for $n=1$, and that $J_{11}(\boldsymbol{k},\boldsymbol{k}')$ is independent of $\boldsymbol{k}$ and $\boldsymbol{k}'$. Then the function $F(\boldsymbol{q},\omega)$ is simplified as

$$F(\boldsymbol{q},\omega)=\frac{2SJ^2}{N}\sum_{k}\left\{\frac{f_{k-q+}-f_{k-}}{\epsilon_{k-q+}-\epsilon_{k-}+\omega}-\frac{f_{k-q+}-f_{k-}}{\epsilon_{k-q+}-\epsilon_{k-}}\right\}, \tag{25}$$

where the suffix 1 has been dropped.

First let us consider the case $\boldsymbol{q}=0$. Then, since

$$F(0,\omega)=\frac{2\sigma J\omega}{\omega-2SJ}, \tag{26}$$

with

$$2N\sigma=\sum_{k}(f_{k+}-f_{k-}),$$

the solution of Eq. (22) can be easily found to be

$$\omega=0, \text{ or } 2J(S+\sigma), \tag{27}$$

if $\omega_{\text{ad}}(0)=0$.

It must be remarked here that there is a solution which does not vanish even if $q=0$. As will be shown in Appendix A by the method of normal modes,[9] this nonvanishing solution corresponds to the oscillation of the system such that band electron spins oscillate in anti-phase with localized spins, which may well be called the *optical mode* of spin waves; while the solution $\omega=0$ corresponds to the *acoustical mode.*

To proceed to the case $\boldsymbol{q}\neq 0$, it is required to calculate Eq. (25), which can be carried out in an elementary way if the following approximations and assumptions are taken:

(1) The energy spectrum of the conduction band is free electron-like, i.e.

$$\epsilon_k=\frac{1}{2}k^2,$$

where the mass of an electron is taken to be unity.

(2) q is so small that it is not required to take into account Umklapp processes.

(3) J/ϵ_F and q/k_F are much smaller than unity, where ϵ_F and k_F denote respectively the Fermi energy and the magnitude of the Fermi wave vector.

Then $F(\boldsymbol{q},\omega)$ is given to the first order of J/ϵ_F and q/k_F as follows:

$$F(\boldsymbol{q},\omega)=F_R(\boldsymbol{q},\omega)+iF_I(\boldsymbol{q},\omega), \tag{28}$$

$$\begin{aligned}F_R(\boldsymbol{q},\omega)=\frac{3SJ^2}{\epsilon_F}\Bigg\{&\frac{(qk_F)^2-\Delta(\Delta-\omega)}{2(qk_F)^2}\\&+\frac{1}{4q^3k_F}\Big([(qk_F-\Delta)(1-q/2k_F)+\omega][(qk_F+\Delta)(1+q/2k_F)-\omega]\\&\qquad\times\ln\left|\frac{(qk_F-\Delta)(1-q/2k_F)+\omega}{(qk_F+\Delta)(1+q/2k_F)-\omega}\right|\\&\quad-[(qk_F-\Delta)(1+q/2k_F)+\omega][(qk_F+\Delta)(1-q/2k_F)-\omega]\\&\qquad\times\ln\left|\frac{(qk_F-\Delta)(1+q/2k_F)+\omega}{(qk_F+\Delta)(1-q/2k_F)-\omega}\right|\Big)\Bigg\},\end{aligned} \tag{29}$$

$$\begin{aligned}F_I(\boldsymbol{q},\omega)=\frac{3SJ^2}{\epsilon_F}\cdot\frac{1}{4q^3k_F}\{&[(qk_F-\Delta)(1-q/2k_F)+\omega][(qk_F+\Delta)(1+q/2k_F)-\omega]\\&\times(\Theta[(qk_F-\Delta)(1-q/2k_F)+\omega]+\Theta[(qk_F+\Delta)(1+q/2k_F)-\omega])\\&-[(qk_F-\Delta)(1+q/2k_F)+\omega][(qk_F+\Delta)(1-q/2k_F)-\omega]\\&\times(\Theta[(qk_F-\Delta)(1+q/2k_F)+\omega]+\Theta[(qk_F+\Delta)(1-q/2k_F)-\omega])\},\end{aligned} \tag{30}$$

with $$\Theta[\epsilon]=\begin{cases}\pi/2 \text{ for } \epsilon>0,\\ -\pi/2 \text{ for } \epsilon<0,\end{cases} \quad \text{and } \Delta=2SJ. \tag{31}$$

For a while we ignore the imaginary part $F_I(\boldsymbol{q}, \omega)$, which causes the damping of spin waves and will be discussed later. When $|\omega-\Delta\pm qk_F|$ is not so small, Eq. (29) can be expanded with $(q/2k_F)\cdot(\Delta\pm qk_F)$ and is reduced to

$$F_R(\boldsymbol{q}, \omega)\cong\frac{3SJ^2}{\epsilon_F}\cdot\frac{\omega}{2qk_F}\ln\left|\frac{qk_F-\Delta+\omega}{qk_F+\Delta-\omega}\right|. \tag{32}$$

When $\omega\cong\Delta-qk_F$,

$$F_R(\boldsymbol{q}, \omega)\cong\frac{3SJ^2}{\epsilon_F}\cdot\frac{\Delta-qk_F}{2qk_F}\ln\left|\frac{\Delta-qk_F}{8e\epsilon_F}\right|, \tag{33}$$

and when $\omega\cong\Delta+qk_F$,

$$F_R(\boldsymbol{q}, \omega)\cong-\frac{3SJ^2}{\epsilon_F}\cdot\frac{\Delta+qk_F}{2qk_F}\ln\left|\frac{\Delta+qk_F}{8e\epsilon_F}\right|. \tag{34}$$

Making use of the above expressions for $F_R(\boldsymbol{q}, \omega)$, its ω dependence is shown in Fig. 1.

For ω being small compared with $\Delta\pm qk_F$, ω can be ignored in the logarithmic function of Eq. (32), and then the solution of Eq. (22) is found easily to be

$$\omega\cong\omega_{\text{ad}}(\boldsymbol{q})\left[1-\frac{3J}{2\epsilon_F}\cdot\frac{\Delta}{2qk_F}\ln\left|\frac{\Delta-qk_F}{\Delta+qk_F}\right|\right]^{-1}, \tag{35}$$

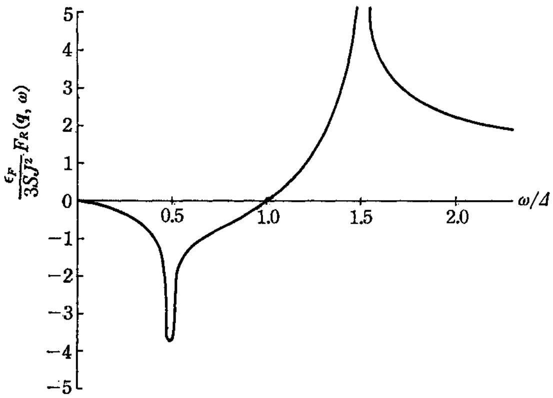

Fig. 1. $F_R(\boldsymbol{q}, \omega)$ as a function of ω for the case $qk_F/\Delta=0.5$ and $J/\epsilon_F=1/5$.

from which we can see that non-adiabatic effects have only a small contribution in this case. As long as ω is not in the vicinity of $|\Delta\pm qk_F|$, the magnitude of the factor $(\Delta/2qk_F)\cdot\ln|(qk_F-\Delta+\omega)/(qk_F+\Delta-\omega)|$ of the expression (32) is of the order unity. Therefore non-adiabatic effects on the spectrum of spin waves can be neglected except for $\omega\cong|\Delta\pm qk_F|$.

Now let us solve Eq. (22) graphically, noting especially the behaviour of $F_R(\boldsymbol{q}, \omega)$ in the vicinity of $\omega\cong|\Delta\pm qk_F|$. In Fig. 2 some typical cases are shown schematically. Then the spectrum of spin waves is given as shown qualitatively in Fig. 3. The figure is rather complicated, and we shall

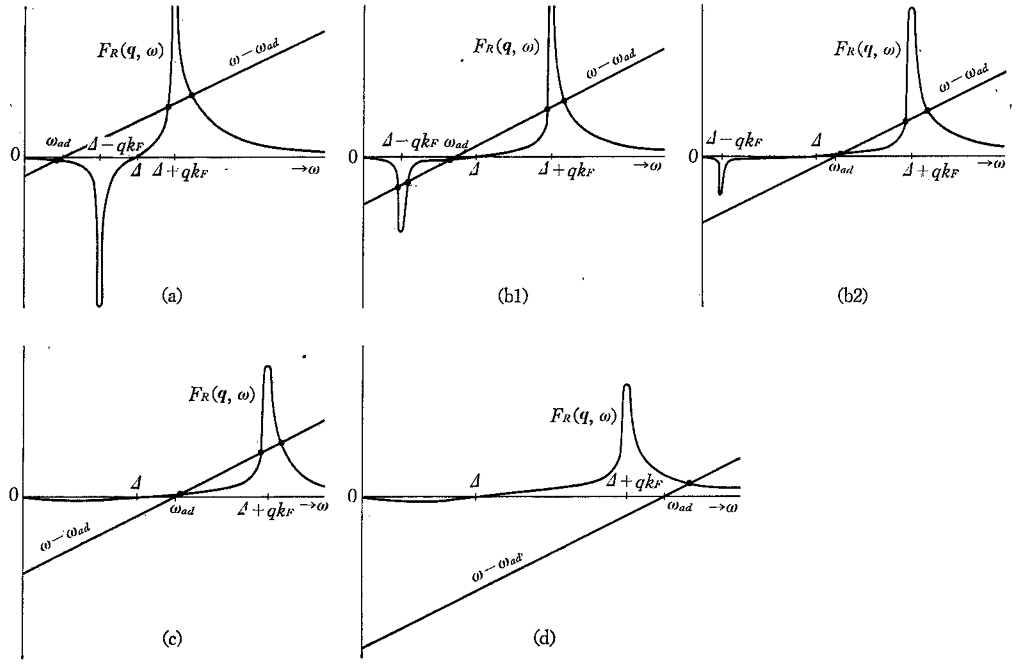

Fig. 2. The equation $\omega-\omega_{ad}=F_R(\boldsymbol{q}, \omega)$ is solved graphically for some typical values of q: (a) $\omega_{ad}<\varDelta-qk_F$, (b) $\varDelta+qk_F>\omega_{ad}>\varDelta-qk_F>0$, (c) $\varDelta-qk_F<0$ and $\omega_{ad}<\varDelta+qk_F$, (d) $\omega_{ad}>\varDelta+qk_F$.

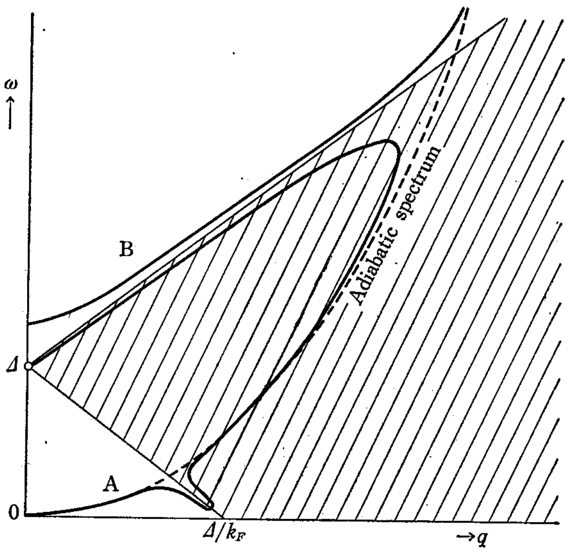

Fig. 3. The spectrum of spin waves obtained by making use of Fig. 2. The dashed line denotes the adiabatic spectrum. In the shaded region spin waves have a finite lifetime. The figure has only a qualitative meaning.

mention some features of the spectrum and give some additional remarks:

(1) When $\omega_{ad}(\boldsymbol{q})\gtrsim\varDelta+qk_F$, there is only one solution and the difference between the acoustical mode and the optical one disappears.

(2) On the other hand, when $\omega_{ad}(\boldsymbol{q})<\varDelta+qk_F$, there appear three or more solutions except for the case $\boldsymbol{q}=0$. Among them, the mode A corresponds to the acoustical one which reduces to zero when $\boldsymbol{q}=0$, and the mode B corresponds to the optical one which reduces to $2J(S+\sigma)$ when $\boldsymbol{q}=0$.

(3) The spectrum of acoustical spin waves are nearly equal

to the adiabatic spectrum except for $\omega_{\rm ad}(\boldsymbol{q})\cong\Delta-qk_F$, as discussed above. On the other hand, when $\omega_{\rm ad}(\boldsymbol{q})\cong\Delta-qk_F$, the spectrum is approximately given by

$$\omega(\boldsymbol{q})\cong\Delta-qk_F. \tag{36}$$

(4) As will be shown in Appendix B, the condition that there appears, in an appreciable degree, the dip discussed above in the spectrum of acoustical spin waves is given by

$$\omega_{\rm ad}(\Delta/k_F)\lesssim 8\epsilon_F\exp\left(1-\frac{4\epsilon_F}{3J}\right). \tag{37}$$

It must be noted here that, though we have shown in Fig. 3 the spectrum in the entire region of $\boldsymbol{q}$, the figure has no meaning where q is too large, since the approximations such that $q/k_F\ll 1$ may not be allowed there. Further discussions on the spectrum will be given in § 5.

§ 4. Damping of spin waves

In § 3 we ignored the imaginary part of the function $F(\boldsymbol{q},\omega)$, and obtained the spectrum of spin waves. When the imaginary part is taken into account, the solution of Eq. (22) becomes complex, and there arises the damping of spin waves. If the imaginary part of the solution is small compared with the real part, the lifetime τ of the spin wave with frequency ω and wave vector $\boldsymbol{q}$ is approximately given by

$$\frac{1}{\tau}=F_I(\boldsymbol{q},\omega),$$

the calculation of which is carried out by the use of Eq. (30). We get

$$\frac{1}{\tau}\cong\frac{3SJ^2}{\epsilon_F}\cdot\frac{\pi}{2qk_F}\times\begin{cases}0 & \text{for } \omega<\Delta-qk_F \text{ and } \omega>\Delta+qk_F,\\ \frac{1}{2}(\Delta-qk_F) & \text{for } \omega\simeq\Delta-qk_F,\\ \frac{1}{2}(\Delta+qk_F) & \text{for } \omega\simeq\Delta+qk_F,\\ \omega & \text{for } \Delta-qk_F<\omega<\Delta+qk_F.\end{cases} \tag{38}$$

This result is essentially equal to that obtained by Mitchell,[10] except that, since we added the diagonal part of *s-d* exchange interaction to one-electron energy of band electrons (see Eq. (15)), the damping is non-vanishing only for $\Delta-qk_F\lesssim\omega\lesssim\Delta+qk_F$: i.e. in the continuum of Stoner's excitations which is shown in Fig. 3 as the shaded region.

Of cource, the approximation taken here is incomplete for the discussion of the damping. At finite temperatures, the damping actually does not vanish even for $\omega < \Delta - qk_F$, because of the fluctuation of the motion of conduction electrons and localized spins. Such effects can be taken into account by the appropriate treatment of higher order Green's functions. We shall not, however, discuss this problem further.

§ 5. Discussions

In the preceding sections, we have investigated non-adiabatic effects on the spectrum of spin waves and shown that there arise mainly two effects. One of them is a dip which appears in the spectrum of ordinary (or acoustical) spin waves when $\omega_{\mathrm{ad}}(\boldsymbol{q}) \simeq 2SJ - qk_F$. Such a deviation from the adiabatic spectrum is appreciable if the condition (37) is satisfied. Supposing that there is no anisotropy energy and that interband terms can be ignored in the expression (23), $\omega_{\mathrm{ad}}(\boldsymbol{q})$ is approximately given by

$$\begin{aligned} \omega_{\mathrm{ad}}(\boldsymbol{q}) &= \frac{J}{N}\sum_{\boldsymbol{k}}\left\{(f_{\boldsymbol{k}+} - f_{\boldsymbol{k}-}) - 2SJ\frac{f_{\boldsymbol{k}-\boldsymbol{q}+} - f_{\boldsymbol{k}-}}{\epsilon_{\boldsymbol{k}-\boldsymbol{q}+} - \epsilon_{\boldsymbol{k}-}}\right\} \\ &\cong \frac{SJ^2}{8\epsilon_F}\left(\frac{q}{k_F}\right)^2, \end{aligned} \tag{39}$$

where $J(\boldsymbol{k}, \boldsymbol{k}')$ is assumed to be independent of $\boldsymbol{k}$ and $\boldsymbol{k}'$, and q/k_F to be much smaller than unity. In this case the condition (37) becomes

$$\left(\frac{J}{\epsilon_F}\right)^4 \lesssim \frac{64e}{S^3}\exp\left(-\frac{4\epsilon_F}{3J}\right), \tag{40}$$

which is fulfilled for $J/\epsilon_F \gtrsim 0.1$ if $S=1$. Usually the magnitude of J is less than 1 eV and that of ϵ_F is several eV in real metals. Therefore it is rather questionable to expect the above condition to be realized literally. However, if the conduction band is anisotropic, there is a possibility that it is satisfied in a certain direction of $\boldsymbol{q}$, and that the above effect appears in that direction. Even if it is satisfied and a dip appears, the number of states of spin waves in the vicinity of the dip is so small that it can hardly contribute to thermal properties of the system.

A more remarkable effect is that if $\omega_{\mathrm{ad}} \lesssim 2SJ + qk_F$ there appear one or more modes of spin waves besides the ordinary one, for instance, an optical mode. The frequency of these excitations is too high, and so they also have no contribution to thermal properties. If they are observed experimentally, it is very interesting because it proposes a way to determine the magnitude of *s-d* exchange interaction in metals.

Finally we must mention the effect of the damping of conduction electrons. If conduction electron spins have a finite lifetime $1/\gamma$, it is shown from a phenomenological argument that the function $F(\boldsymbol{q}, \omega)$ given by Eq. (25) must be replaced by

$$F_\gamma(\boldsymbol{q}, \omega) = \frac{2SJ^2}{N}\sum_k \left\{ \left(1 - i\frac{\gamma}{\Delta}\right) \frac{f_{k-q+} - f_{k-}}{\epsilon_{k-q+} - \epsilon_{k-} + \omega + i\gamma} - \frac{f_{k-q+} - f_{k-}}{\epsilon_{k-q+} - \epsilon_{k-}} \right\}. \tag{41}$$

The factor $(1-i\gamma/\Delta)$ of the first term in the wavy brackets comes from the fact that the density of conduction electron spins relaxes towards the local equilibrium value. If $\gamma \ll \Delta$, the real part of the function $F_\gamma(\boldsymbol{q}, \omega)$ is nearly equal to $F_R(\boldsymbol{q}, \omega)$ except for $\omega \simeq \Delta \pm qk_F$. Therefore most of the preceding arguments need not be modified but for the condition (37), i.e. the depth of the dip will be smaller than considered above. As to the imaginary part, there arises also a weak damping of spin waves from γ even when $\omega < |\Delta - qk_F|$ and $\omega > |\Delta + qk_F|$, i.e. for $\omega \ll |\Delta - qk_F|$,

$$\frac{1}{\tau} = \frac{\gamma}{8S^2J} q^2, \tag{42}$$

which is much smaller than the damping for $|\Delta - qk_F| < \omega < |\Delta + qk_F|$. After all, it can be concluded that effects of γ are not qualitative, but only quantitative ones, and that important modifications are not necessary.

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Appendix A

Method of normal modes

In this appendix we shall introduce briefly the method of normal modes given by Matsubara[9] and investigate to what normal modes the spin wave excitations discussed in § 3 correspond.

Let us take normal modes of the system as

$$Q^*(\boldsymbol{q}) = \sum_k c_k\, a_{k-}^*\, a_{k-q+} + gS^-(-\boldsymbol{q}), \tag{A·1}$$

where c_k and g are constants which are to be determined later. Here interband terms have been ignored for simplicity. Then we have the relation

$$[H, Q^*(\boldsymbol{q})]\,|0\rangle = \omega|\boldsymbol{q}\rangle, \tag{A·2}$$

where $|0\rangle$ denotes the ground state of the system and ω is the excitation energy of the state $|\boldsymbol{q}\rangle = Q^*(\boldsymbol{q})\,|0\rangle$.

Using a simplified Hamiltonian where interband *s-d* interactions are omitted at the outset, and taking the approximation

$$\begin{aligned} S^0(\boldsymbol{k})\,|0\rangle &= \delta_{k0} NS|0\rangle, \\ a_{k\sigma}^*\, a_{k'\sigma}|0\rangle &= \delta_{kk'} f_{k\sigma}|0\rangle, \end{aligned} \tag{A·3}$$

for the ground state, we get from Eq. (A·2) the following relation:

$$\sum_k \{(\epsilon_{k-}-\epsilon_{k-q+}-\omega)c_k - g2SJ(\boldsymbol{k}-\boldsymbol{q},\boldsymbol{k})\}a_{k-}\,a_{k-q+}|0\rangle$$

$$+\{g(\Delta-\omega)-\frac{1}{N}\sum_k J(\boldsymbol{k},\boldsymbol{k}-\boldsymbol{q})(c_{k+q}f_{k+}-c_k f_{k-})\}S^-(-\boldsymbol{q})|0\rangle=0. \quad (\text{A}\cdot 4)$$

It is noted here that the approximation (A·3) is equivalent to that taken in § 2 for higher order Green's functions (cf. Eq. (10)). From Eq. (A·4) the constants c_k's are determined as

$$c_k=-g\frac{2SJ(\boldsymbol{k}-\boldsymbol{q},\boldsymbol{k})}{\omega-(\epsilon_{k-}-\epsilon_{k-q+})}, \quad (\text{A}\cdot 5)$$

and the energy ω is to be obtained from the relation

$$\omega=\Delta+\frac{2S}{N}\sum_k |J(\boldsymbol{k},\boldsymbol{k}-\boldsymbol{q})|^2\frac{f_{k-q+}-f_{k-}}{\omega-(\epsilon_{k-}-\epsilon_{k-q+})}, \quad (\text{A}\cdot 6)$$

which is equal to Eq. (22) in the text if interband terms are ignored there.

As was shown in § 3, Eq. (A·6) has two solutions $\omega=0$ and $\omega=2J(S+\sigma)$ for the case $\boldsymbol{q}=0$. Making use of Eq. (A·5), the normal mode for each solution is found to be

$$Q_I^*(0)=g[\sum_k a_{k-}^*\,a_{k+}+S^-(0)] \qquad \text{for } \omega=0,$$

$$Q_{II}^*(0)=g[-(S/\sigma)\sum_k a_{k-}^*\,a_{k+}+S^-(0)] \quad \text{for } \omega=2J(S+\sigma). \quad (\text{A}\cdot 7)$$

Now it is clear from these expressions that Q_I is the acoustical mode where two spin systems, localized spins and conduction electron spins, oscillate in phase, while Q_{II} is the optical mode where they oscillate in antiphase with each other.

It is interesting to show the expressions of the normal modes for $\omega=\Delta\pm qk_F+\varepsilon$, ε being a small positive or negative quantity, where the spectrum deviates from the adiabatic one. We get

$$Q^*(\boldsymbol{q})\cong g[S^-(-\boldsymbol{q})\pm 2SJ\sum_k\frac{a_{k-}^*\,a_{k-q+}}{qk_F(1\pm\cos\theta)+\varepsilon}], \quad (\text{A}\cdot 7')$$

where θ is the angle between $\boldsymbol{k}$ and $\boldsymbol{q}$. From Eq. (A·7′) these excitations can be understood essentially as the spin waves of localized spins which accompany the Stoner excitations $a_{k-}^* a_{k-q+}$ of conduction electrons such that $\boldsymbol{k}$ is parallel ($\cos\theta=1$) or antiparallel ($\cos\theta=-1$) to $\boldsymbol{q}$.

Appendix B

Condition for the appreciable appearance of a dip in the spectrum of acoustical spin waves

As is shown in Fig. 1, $F_R(\boldsymbol{q},\omega)$, as a function of ω, has a sharp minimum at $\omega=\Delta-qk_F$. Because of this feature of $F_R(\boldsymbol{q}, \omega)$, the solution of Eq. (22) deviates from the adiabatic value $\omega=\omega_{\rm ad}(\boldsymbol{q})$ when $\omega_{\rm ad}\simeq\Delta-qk_F$. At the same time it is noted that the absolute value of the minimum decreases as $(\Delta-qk_F)$ decreases, and that, if the inequality

$$|\omega_{\rm ad}(\boldsymbol{q})-(\Delta-qk_F)|<\frac{3SJ^2}{\epsilon_F}\cdot\frac{\Delta-qk_F}{2qk_F}\ln\left(\frac{8e\epsilon_F}{\Delta-qk_F}\right) \tag{A·8}$$

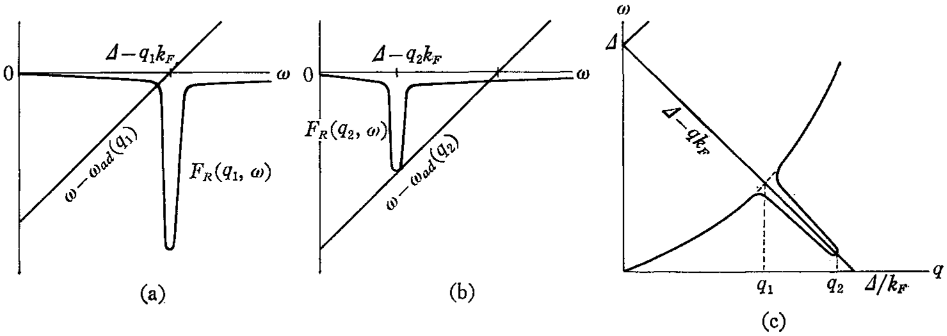

Fig. 4. In the region between q_1 and q_2, which are determined respectively by (a) and (b), the spectrum of spin waves deviates from the adiabatic one as is shown in (c).

is not satisfied, the solution of Eq. (22) is again given by $\omega\simeq\omega_{\rm ad}(\boldsymbol{q})$. The situation is shown schematically in Fig. 4, i.e. when q exceeds q_1 determined by

$$\omega_{\rm ad}(q_1)=\Delta-q_1k_F \tag{A·9}$$

(Fig. 4 (a)), the spectrum of acoustical spin waves begins to deviate from the adiabatic one, and when q exceeds q_2 determined by

$$|\omega_{\rm ad}(q_2)-(\Delta-q_2k_F)|=\frac{3SJ^2}{\epsilon_F}\cdot\frac{\Delta-q_2k_F}{2q_2k_F}\ln\left(\frac{8e\epsilon_F}{\Delta-q_2k_F}\right) \tag{A·10}$$

(Fig. 4 (b)), the spectrum returns again to the adiabatic one. Then, as is seen from Fig. 4 (c), the condition that a dip appears appreciably in the spectrum is given by

$$\frac{q_2-q_1}{\Delta/k_F-q_1}=O(1). \tag{A·11}$$

Introducing dimensionless parameters defined by

$$\left.\begin{aligned}&\frac{\Delta-q_1k_F}{8e\epsilon_F}=x_1,\quad \frac{\Delta-q_2k_F}{8e\epsilon_F}=x_2,\\ &\frac{\omega_{\rm ad}(\Delta/k_F)}{8e\epsilon_F}=\alpha,\quad \frac{3J}{4\epsilon_F}=\beta,\end{aligned}\right\} \tag{A·12}$$

and taking the lowest approximation of small quantities x_1 and x_2, we can reduce Eqs. (A·9), (A·10) and (A·11) to

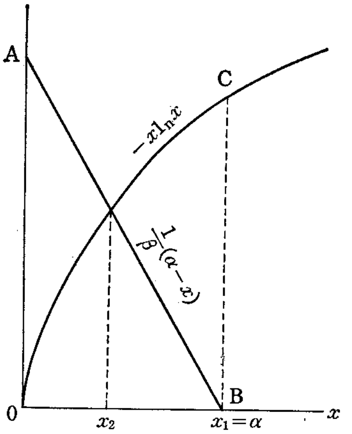

Fig. 5. Eqs. (A·13) and (A·14) are solved graphically.

$$\alpha-x_1=0, \tag{A·13}$$

$$\frac{1}{\beta}(\alpha-x_2)=-x_2\ln x_2, \tag{A·14}$$

$$\frac{x_1-x_2}{x_1}=O(1). \tag{A·15}$$

Equation (A·14) is solved graphically in Fig. 5, and the condition (A·15) becomes $\overline{OA}\lesssim\overline{BC}$ in this figure. Therefore we get the condition for the appreciable appearance of a dip in the spectrum as

$$\alpha\lesssim\exp\left(-\frac{1}{\beta}\right), \tag{A·16}$$

or making use of the relations (A·12),

$$\omega_{\text{ad}}(\varDelta/k_F)\lesssim 8e\epsilon_F\exp\left(-\frac{4\epsilon_F}{3J}\right). \tag{A·17}$$

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