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SPIN POLARIZATION OF A MAGNETIC ELECTRON
GAS INDUCED BY A VAN VLECK ION

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Spin Polarization of a Magnetic Electron Gas Induced by a
van Vleck Ion.

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ABSTRACT

The mutual polarization of a magnetic electron gas and a van Vleck ion, interacting via exchange, are theoretically investigated using the double-time Green function method. A pair of equations describing the dynamics of the electron gas and the ion are conveniently decoupled and an analytic expression for the electron gas polarization, which depends on the square of the exchange parameter, is obtained. Besides a RKKY-like term, a new term associated to the process of formation of the magnetic moment of the ion appears.

Les polarisations mutuelles d'un gaz électronique magnétisé et d'un ion van Vleck, interagissant par échange, sont étudiées théoriquement en utilisant la méthode de la fonction de Green à double temps. Les paires d'équations décrivant la dynamique du gaz électronique et de l'ion sont découplées. On obtient une expression analytique de la polarisation du gaz électronique, dépendant du carré du paramètre d'échange. Outre un terme du type RKKY, il apparaît un nouveau terme associé au processus de formation du moment magnétique de l'ion.

1. INTRODUCTION

The phenomenon of spin polarization of an electron gas due to exchange interaction with an ion which has a local spin, the so called RKKY (Ruderman-Kittel-Kasuya-Yosida) effect, is a classical subject in solid state physics [1]. Through this polarization, which is of long range character and oscillatory, localized magnetic moments of rare-earth ions in metals and alloys can be indirectly coupled giving rise to complex magnetic structures [2,3].

The idea of spin polarization and indirect coupling have been generalized by several authors. Bloembergen and Rowland have applied the concept of indirect coupling between localized spins to insulators; here the interaction is realized by virtual excitations through the energy gap of the valence band [4]. Blandin and Campbell have extended the RKKY effect to an electron gas strongly perturbed by an electric charge in connection with the problem of the magnetism of Heusler's alloys [5]. Also the polarization effect due to a local spin on an already magnetized band was studied by Chatel and Szabo [6].

In this paper we consider the problem of how a van Vleck ion can polarize an electron gas and if there is any significant difference to the usual RKKY polarization. We begin by assuming a magnetic conduction band which interacts by exchange with a van Vleck ion subjected to a crystalline field. Actually we have a problem of mutual magnetization; the magnetic band induces a magnetization on the ion which responds polarizing

the electron gas in a self-consistent way. A spin polarization induced by a van Vleck ion was proposed by Bloch et al. [7] in connection with EPR measurements of Gd diluted in intermetallic compounds MBe_{13} (where M is Pr, Eu, Tm and U). These systems, according to Bucher et al. [8], exhibit van Vleck paramagnetism. In [7] in order to explain g-shifts of Gd diluted in MBe_{13} , in addition to the local exchange interaction with the conduction electrons, it was also necessary to assume an indirect exchange coupling, RKKY-like, between the Gd ion and the surrounding van Vleck ions.

The plan of the paper is as follows. In the next section we introduce the model Hamiltonian and specify the quantities to be computed. In the section 3 we define the appropriate Green's functions (GF), introduce the necessary approximations and obtain the GF associated to the electron gas polarization, In section 4 we evaluate the statistical averages of the pseudo-spins relative to the van Vleck ion. Finally, in section 5, we obtain an expression for the electronic polarization as a function of the distance from the ion and discuss the physical meaning of the several contributions.

2. FORMULATION OF THE PROBLEM

Let us consider a rare-earth ion embedded in a metallic matrix having a magnetic band. A crystalline field splits the ion energy level in only two levels which are supposed to be not degenerated. The energy difference between these two singlet levels is Δ . Taking into account the exchange interaction of this

van Vleck ion with the conduction electrons the system can be described by the following model Hamiltonian

$$H = \sum_{\vec{k}\sigma} \epsilon_{\vec{k}\sigma} c_{\vec{k}\sigma}^+ c_{\vec{k}\sigma} - \Delta S^x - J s^z S^z \quad (1)$$

J is an effective exchange integral, namely

$$J = 4 J' (g-1)M \quad (2)$$

where M is the matrix element of the angular momentum between the two levels of the ion, g the Lande's factor and J' is the exchange integral of the localized and itinerant electrons. S^x and S^z represent pseudo-spin operators associated to the ion and

$$s^z = \frac{1}{2N} \sum_{\vec{k}\vec{k}'} (c_{\vec{k}'\uparrow}^+ c_{\vec{k}\uparrow} - c_{\vec{k}'\downarrow}^+ c_{\vec{k}\downarrow}) \quad (3)$$

is the spin polarization of the conduction electrons, where $c_{\vec{k}\sigma}^+$ ($c_{\vec{k}\sigma}$) is the creation (annihilation) operator associated to an electron of wave number \vec{k} and spin σ ($\sigma = \pm 1$, corresponding to up and down).

This model Hamiltonian was introduced by Fulde and Peschel [9] and Schotte [10] who have studied the dynamic susceptibility of a van Vleck ion interacting with an electron gas. In order to emphasize the different origins of the pseudo-spins S^x , S^z and the electron spin s^z , in appendix A, (1) is derived from a more naive version.

In this work we want to determine the spin polarization of an electron gas induced by a van Vleck ion put at the origin in function of the distance. The quantity of interest is

$$s^z(\mathbf{r}) = \frac{1}{2N} \sum_{\mathbf{q}} e^{i\mathbf{q} \cdot \mathbf{r}} \sum_{\mathbf{k}} \langle (c_{\mathbf{k}+\mathbf{q}\uparrow}^+ c_{\mathbf{k}\uparrow} - c_{\mathbf{k}+\mathbf{q}\downarrow}^+ c_{\mathbf{k}\downarrow}) \rangle \quad (4)$$

In evaluating $\langle c_{\mathbf{k}'\sigma}^+ c_{\mathbf{k}\sigma} \rangle$ we adopt a perturbative scheme in the parameter J . As we will see, in the next section, the polarization $s^z(\mathbf{r})$ involves the quantities $\langle S^\pm \rangle$ and $\langle S^z \rangle$; these are also computed in the first order in J (section 4).

3. Green Function of the electronic spin polarization in a perturbative scheme

In what follows we use the Zubarev's algorithm which allows to compute the average $\langle BA \rangle$ from the double time GF $\langle A|B \rangle$

$$\langle BA \rangle = i \lim_{\epsilon \rightarrow 0} \int d\omega \frac{\langle A|B \rangle_{\omega+i\epsilon} - \langle A|B \rangle_{\omega-i\epsilon}}{e^{\beta\omega} \pm 1} \quad (5)$$

$\langle A|B \rangle$ obeys

$$\omega \langle A|B \rangle_{\omega} = \frac{1}{2\pi} \langle [A, B]_{\pm} \rangle + \langle [A, H]_{-} | B \rangle_{\omega} \quad (6)$$

The statistical averages which appear in (5) and (6) are defined in a canonical ensemble. If a grand canonical is preferred one should replace in (6) H for $H' = H - \mu N$ where μ is the chemical potential and N the number operator [12].

In order to compute (4) we introduce the GF $\langle c_{\mathbf{k}\sigma} | c_{\mathbf{k}'\sigma}^+ \rangle$.

We will need the following results

$$[c_{k\sigma}, H']_- = (\epsilon_{k\sigma} - \mu) c_{k\sigma} - \frac{J\sigma}{N} \sum_k S^Z c_{k\sigma} \quad (7-a)$$

$$[S^Z c_{k\sigma}, H']_- = (\epsilon_{k\sigma} - \mu) S^Z c_{k\sigma} - \frac{\Delta}{2} (S^+ - S^-) c_{k\sigma} \quad (7-b)$$

Since we intend to compute $\langle c_{k\sigma} | c_{k'\sigma}^+ \rangle$ until second order in J we define $\langle c_{k\sigma} | c_{k'\sigma}^+ \rangle^{(i)}$, $i = 1, 2$, as the perturbed GF of order i in the parameter J. We also define the zeroth order GF

$$\langle c_{k\sigma} | c_{k'\sigma}^+ \rangle^{(0)} = \frac{\delta_{k,k'}}{2\pi} \frac{1}{\omega - (\epsilon_{k\sigma} - \mu)} \quad (8)$$

Using (7) we obtain, according to (6)

$$[\omega - (\epsilon_{k\sigma} - \mu)] \langle c_{k\sigma} | c_{k'\sigma}^+ \rangle^{(i)} = - \frac{J\sigma}{N} \langle S^Z c_{k\sigma} | c_{k'\sigma}^+ \rangle^{(i-1)} \quad (9)$$

$$[\omega - (\epsilon_{k\sigma} - \mu)] \langle S^Z c_{k\sigma} | c_{k'\sigma}^+ \rangle = \frac{\delta_{k,k'}}{2\pi} \langle S^Z \rangle - \frac{\Delta}{2} \langle (S^+ - S^-) c_{k\sigma} | c_{k'\sigma}^+ \rangle \quad (10)$$

At this point we make the following approximation

$$\langle S^\pm c_{k\sigma} | c_{k'\sigma}^+ \rangle \approx \langle S^\pm \rangle \langle c_{k\sigma} | c_{k'\sigma}^+ \rangle \quad (11)$$

Since $\langle S^Z \rangle^{(0)} = (\langle S^+ \rangle^{(0)} - \langle S^- \rangle^{(0)}) = 0$, see section 4, only $\langle S^Z c_{k\sigma} | c_{k'\sigma}^+ \rangle^{(1)}$ contributes

$$[\omega - (\epsilon_{k\sigma} - \mu)] \langle S^Z c_{k\sigma} | c_{k'\sigma}^+ \rangle^{(1)} = \frac{\delta_{k,k'}}{2\pi} \langle S^Z \rangle^{(1)} - \frac{\Delta}{2} (\langle S^+ \rangle^{(1)} - \langle S^- \rangle^{(1)}) \langle c_{k\sigma} | c_{k'\sigma}^+ \rangle^{(0)} \quad (12)$$

Solving for (8), (9) and (10) we finally obtain

$$\langle c_{k\sigma} | c_{k'\sigma}^+ \rangle^{(2)} = -\frac{J\sigma}{N} \frac{1}{2\pi} \left\{ \frac{\langle S^Z \rangle^{(1)}}{[\omega - (\epsilon_{k'\sigma} - \mu)] [\omega - (\epsilon_{k\sigma} - \mu)]} + \frac{\Delta}{2} \frac{(\langle S^+ \rangle^{(1)} - \langle S^- \rangle^{(1)})}{[\omega - (\epsilon_{k'\sigma} - \mu)]^2 [\omega - (\epsilon_{k\sigma} - \mu)]} \right\} \quad (13)$$

(13) involves the quantities $\langle S^Z \rangle$ and $\langle S^\pm \rangle$ in first order in J. These are computed in the next section.

4. Evaluation of $\langle S^Z \rangle$ and $\langle S^\pm \rangle$

In order to obtain $\langle S^Z \rangle$ and $\langle S^\pm \rangle$ we use the following relations

$$S^Z = \frac{1}{2} - S^- S^+ \quad (14-a)$$

$$S^\pm S^Z = \mp \frac{1}{2} S^\pm \quad (14-b)$$

Similarly to what was done in section 3. We apply (5) to evaluate $\langle S^\pm S^Z \rangle$ and $\langle S^- S^+ \rangle$. We introduce the following GF $\langle S^+ | S^- \rangle$ and $\langle S^Z | S^+ \rangle$.

The application of GF technique to the computation of statistical averages involving pseudo-spins was done by Ramakrishnan et al. [13] in the case of ferroelectric Hamiltonian of the kind order-disorder. As in [13] we work with the anticommutator.

The following results will be used

$$[S^\pm, H]_- = \pm 2J s^Z S^\pm \mp \Delta S^Z \quad (15-a)$$

$$[\bar{S}^Z, H]_- = -\frac{\Delta}{2} (S^+ - S^-) \quad (15-b)$$

Similarly to (11) we also put

$$\langle s^Z S^\pm | S^\pm \rangle = \langle s^Z \rangle \langle S^\pm | S^\pm \rangle \quad (16)$$

We then obtain

$$\omega \langle S^+ | S^- \rangle = \frac{1}{2\pi} + 2J \langle s^Z \rangle \langle S^+ | S^- \rangle - \Delta \langle S^Z | S^- \rangle \quad (17-a)$$

$$\omega \langle S^Z | S^\pm \rangle = -\frac{\Delta}{2} (\langle S^+ | S^\pm \rangle - \langle S^- | S^\pm \rangle) \quad (17-b)$$

$$\omega \langle S^+ | S^+ \rangle = 2J \langle s^Z \rangle \langle S^+ | S^+ \rangle - \Delta \langle S^Z | S^+ \rangle \quad (17-c)$$

$$\omega \langle S^- | S^- \rangle = -2J \langle s^Z \rangle \langle S^- | S^- \rangle + \Delta \langle S^Z | S^- \rangle \quad (17-d)$$

$$\omega \langle S^- | S^+ \rangle = \frac{1}{2\pi} - 2J \langle s^Z \rangle \langle S^- | S^+ \rangle + \Delta \langle S^Z | S^+ \rangle \quad (17-e)$$

After a tedious but simple algebra we obtain

$$\langle S^+ | S^- \rangle = \frac{1}{2\pi} \frac{2\omega^2 + 4J \langle s^Z \rangle \omega - \Delta^2}{2\omega [\omega^2 - (\Delta^2 + 4J^2 \langle s^Z \rangle^2)]} \quad (18-a)$$

$$\langle S^Z | S^- \rangle = -\frac{1}{2\pi} \frac{\Delta}{2} \frac{(\omega + 2J \langle s^Z \rangle)}{\omega(\omega^2 - 4J^2 \langle s^Z \rangle^2 - \Delta^2)} \quad (18-b)$$

$$\langle S^Z | S^+ \rangle = \frac{1}{2\pi} \frac{\Delta}{2} \frac{\omega - 2J \langle s^Z \rangle}{\omega(\omega^2 - 2J \langle s^Z \rangle \omega - \Delta^2)} \quad (18-c)$$

From (14) and (17) we obtain, for $T = 0$, $\langle S^\pm \rangle$ and $\langle S^Z \rangle$

(see appendix B). In first order in J

$$\langle S^z \rangle = \frac{J}{\Delta} \langle s^z \rangle \quad (19)$$

$$\langle S^+ \rangle - \langle S^- \rangle = -\frac{2J}{\Delta} \langle s^z \rangle \quad (20)$$

5. Electron gas polarization at T=0

The aim of this section is to compute $s^z(r)$ defined in (4). We begin evaluating

$$\sum_{k\sigma} \sigma \langle c_{k+q\sigma}^+ c_{k\sigma} \rangle \quad (21)$$

From (13) and using (5) one obtains

$$\begin{aligned} \sum_{k\sigma} \sigma \langle c_{k+q\sigma}^+ c_{k\sigma} \rangle^{(2)} &= -J \langle S^z \rangle^{(1)} F_1(q) + [F_2(q) + F_3(q)] \\ &\quad \times \frac{\Delta}{2} (\langle S^+ \rangle^{(1)} - \langle S^- \rangle^{(1)}) \end{aligned} \quad (22)$$

where

$$F_1(q) = \sum_{k\sigma} \frac{f(\epsilon_{k\sigma}) - f(\epsilon_{k+q\sigma})}{\epsilon_{k\sigma} - \epsilon_{k+q\sigma}} \quad (23-a)$$

$$F_2(q) = \sum_{k\sigma} \frac{f(\epsilon_{k\sigma}) - f(\epsilon_{k+q\sigma})}{(\epsilon_{k\sigma} - \epsilon_{k+q\sigma})^2} \quad (23-b)$$

$$F_3(q) = - \sum_{k\sigma} \frac{f'(\epsilon_{k+q\sigma})}{(\epsilon_{k\sigma} - \epsilon_{k+q\sigma})^2} \quad (23-c)$$

$F_2(q)$ and $F_3(q)$ come from the second term of (13); $F_1(q)$ comes

from the first one.

Putting

$$\epsilon_{k\sigma} = \epsilon_k - \frac{(\sigma-1)}{2} A \quad (24)$$

where $\sigma = \pm 1$ and A is the shift of the up and down bands.

At $T=0$ and for $\epsilon_k = \frac{(\hbar k)^2}{2m}$, $E_F > A$ (23) become

$$F_1(q) = - \frac{m}{2\pi^2 \hbar^2} \left[k_F g(k_F, q) + k_{F\downarrow} g(k_{F\downarrow}, q) \right] \quad (25)$$

where

$$k_{F\downarrow} = \frac{\sqrt{2m(E_F - A)}}{\hbar}$$

k_F is given by

$$\frac{1}{2} \left[\int_0^{E_F} g(\epsilon) d\epsilon + \int_A^{E_F} g(\epsilon - A) d\epsilon \right] = \eta \quad (26)$$

where

$$g(\epsilon) = \frac{1}{2} \frac{m}{\hbar^2 \pi} \frac{2m}{\hbar^2} \epsilon^{1/2} \epsilon^{1/2}$$

n is the number of electrons per atom and

$$g(k, q) = \frac{1}{2} + \frac{1}{8} \frac{4k^2 - q^2}{kq} \ln \left| \frac{2k+q}{2k-q} \right| \quad (27)$$

$$F_2(q) = 0 \quad (28)$$

and

$$F_3(q) = \frac{m^2}{4\pi^2 \hbar^4 q} \left[h(k_F, q) + h(k_{F\downarrow}, q) \right] \quad (29)$$

where

$$h(k, q) = \lambda n \left| \frac{2k+q}{2k-q} \right| \quad (30)$$

In appendix C more details about (25), (28) and (29) are given.

Expression (4) can be written in the form

$$s^z(r) = s_1^z(r) + s_2^z(r) \quad (31)$$

where

$$s_1^z(r) = - \frac{J \langle S^z \rangle}{N} \sum_q e^{iq \cdot r} F_1(q) \quad (32-a)$$

$$s_2^z(r) = - \frac{J (\langle S^+ \rangle^{(1)} - \langle S^- \rangle^{(1)})}{N} \sum_q e^{iq \cdot r} F_3(q) \quad (32-b)$$

Integrating in q and taking into account (21) and (22) we finally obtain

$$s_1^z(r) = \begin{cases} BF(2k_F r) & \text{if } E_F < 2A \\ E \left[\overline{F}(2k_F r) + \left(\frac{k_{F\downarrow}}{k_F} \right)^2 F(2k_{F\downarrow} r) \right] & \text{if } E_F \geq 2A \end{cases} \quad (33)$$

where

$$F(x) = \frac{\text{sen } x - x \cos x}{x^4}$$

and

$$B = \frac{2J_m^2 \langle s^z \rangle}{\pi N \Delta \hbar^2} k_F^4$$

$$s_2^z(r) = \begin{cases} c \phi(2k_F r) & \text{if } E_F < \Delta \\ c \left[\phi(2k_F r) + \left(\frac{k_{F\downarrow}}{k_F} \right)^2 \phi(2k_{F\downarrow} r) \right] & \text{if } E_F \geq \Delta \end{cases}$$

where

$$\phi(x) = \frac{\text{sen } x}{x^2}$$

and

$$c = \frac{J^2 m \langle s^z \rangle k_F^4}{4 \pi^3 N \hbar^2 E_F}$$

the polarization expression (31) contains two contributions $s_1^z(r)$. RKKY-like, and $s_2^z(r)$. Going back to (13) we see that $s_1^z(r)$ and $s_2^z(r)$ are associated with $\langle S^z \rangle^{(1)}$ and $(\langle S^+ \rangle - \langle S^- \rangle)^{(1)}$ respectively. So $s_1^z(r)$ can be interpreted in the following way: the magnetic band induces a polarization $\langle S^z \rangle^{(1)}$ on the ion which reacts polarizing the electron gas in the usual RKKY way. $s_2^z(r)$, as suggested by the coefficient $(\langle S^+ \rangle - \langle S^- \rangle)^{(1)}$, is associated to the process of formation of the magnetic moment of the ion. Since one has a mutual polarization process the final electronic polarization depends on J^2 . Worthnoting is the Δ dependence of $s_1^z(r)$ but not of $s_2^z(r)$.

APPENDIX A

A two energy singlet ion interacting via local exchange with an electron gas can be described by the following model Hamiltonian

$$H = H_{e\ell} + H_{ion} + H_{e\ell-ion} \quad (A-1)$$

where

$$H_{e\ell} = \sum_{k\sigma} \epsilon_{k\sigma} c_{k\sigma}^{\dagger} c_{k\sigma} \quad (\text{A-2})$$

$$H_{\text{ion}} = \begin{pmatrix} 0 & 0 \\ 0 & \Delta \end{pmatrix} \quad (\text{A-3})$$

$$H_{e\ell\text{-ion}} = J'(g-1) s^z \begin{pmatrix} \langle 0|S^z|0\rangle & \langle 0|S^z|e\rangle \\ \langle e|S^z|0\rangle & \langle e|S^z|e\rangle \end{pmatrix}$$

where J' is the exchange integral and g the Lande's factor.

For a van Vleck ion $\langle 0|S^z|0\rangle = \langle e|S^z|e\rangle = 0$;

$\langle 0|S^z|e\rangle = \langle e|S^z|0\rangle = M \neq 0$.

So

$$H_{e\ell\text{-ion}} = J'M(g-1) s^z \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad (\text{A-4})$$

Transforming from $|0\rangle, |e\rangle$ to $|\bar{0}\rangle, |\bar{e}\rangle$ by

$$\begin{pmatrix} |\bar{0}\rangle \\ |\bar{e}\rangle \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix} \begin{pmatrix} |0\rangle \\ |e\rangle \end{pmatrix} \quad (\text{A-5})$$

one obtains

$$\bar{H} = H_{e\ell} - \Delta S^x - J s^z S^z \quad (\text{A-6})$$

where $J = 4J'M(g-1)$ and S^x, S^z are pseudo-spin operators, namely

$$S^x = \frac{1}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}; \quad S^z = \frac{1}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

APPENDIX B

Using (18), (14) and applying (5) one obtains for
T=0

$$\langle S^z \rangle = \frac{J \langle S^z \rangle \sqrt{4J^2 \langle S^z \rangle^2 + \Delta^2}^{1/2}}{2(\Delta^2 + 4J^2 \langle S^z \rangle^2)} \quad (B-1)$$

$$\langle S^- \rangle = \Delta \frac{4J \langle S^z \rangle + \sqrt{4J^2 \langle S^z \rangle^2 + \Delta^2}^{1/2}}{2(4J^2 \langle S^z \rangle^2 + \Delta^2)} \quad (B-2)$$

$$\langle S^+ \rangle = -\Delta \left[\frac{2J \langle S^z \rangle}{\sqrt{4J^2 \langle S^z \rangle^2 + \Delta^2} + J \langle S^z \rangle} \sqrt{4J^2 \langle S^z \rangle^2 + \Delta^2}^{1/2} - \frac{(4J^2 \langle S^z \rangle^2 + \Delta^2)^{1/2}}{4J^2 \langle S^z \rangle^2 + \Delta^2} \frac{J \langle S^z \rangle \sqrt{4J^2 \langle S^z \rangle^2 + \Delta^2}^{1/2}}{4} \right] \quad (B-3)$$

Expanding in first order in J one obtains (19) and (20).

APPENDIX C

Summing for σ in (23-a), using (24) and following [1] we obtain (25).

At T=0 (23-b) can be written

$$F_2(q) = \sum_{\sigma} k^2 dk \left[\int_{-1}^{+1} \frac{d\mu}{(q+2k\mu)^2} - \int_{-1}^{+1} \frac{d\mu}{(q-2k\mu)^2} \right] \quad (C-1)$$

where $\mu = \cos\theta$, θ is the angle between \vec{k} and \vec{q} . The term in brackets

is obviously zero.

Integrating (23-c) in spherical coordinates in the \vec{k} space and summing in σ we obtain

$$F_3(q) = - \frac{m}{4\pi^2 \hbar^2 q} \left\{ \int_0^{k_F} dk \delta(\epsilon_k - \epsilon_F) k \ln \left| \frac{2k+q}{2k-q} \right| \right. \\ \left. + \int_0^{k_{F\downarrow}} dk \delta[\epsilon_k - (\epsilon_F - A)] k \ln \left| \frac{2k+q}{2k-q} \right| \right\} \quad (C-2)$$

From C-2, for $\epsilon_k = \frac{(\hbar k)^2}{2m}$ we obtain (29).

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