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Autor(en): **Baltensperger, W. / Graaf, A.M. de**

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Long range interactions between magnetic moments in semiconductors

by **W. Baltensperger** and **A. M. de Graaf**

(Laboratorium für Festkörperphysik, ETH, Zürich)

Summary: The interaction between magnetic moments embedded in a non degenerate electron gas is calculated. In the fully degenerate gas the RUDERMAN, KITTEL interaction applies whereas in the Boltzmann gas the interaction is ferromagnetic only and of comparatively long range. Between these extreme cases the behaviour changes gradually. In a semiconductor an additional interaction comes from the virtual excitations of the valence band. These two interactions and the magnetic dipole interaction may be of comparable magnitude in dilute magnetic semiconducting alloys.

1. Introduction

PAULI¹⁾ treated the spin susceptibility of a degenerate free electron gas for the case of a uniform external magnetic field. His work has been extended to finite temperatures²⁾. The space dependence of the spin polarisation due to a localized magnetic field has been calculated by RUDERMAN, KITTEL³⁾ and YOSIDA⁴⁾. In the present paper this will be considered for a non degenerate electron gas.

Localized actions on the spins of conduction electrons are caused by the electron exchange with magnetic ions or by the hyperfine interaction with magnetic nuclei. The resulting polarisation of the electron gas leads to an interaction between magnetic ions or nuclei. Such interactions were described by FRÖHLICH, NABARRO⁵⁾, VONSOVSKII⁶⁾, RAMSEY, PURCELL⁷⁾ and RUDERMAN, KITTEL³⁾. In a semiconductor the non degenerate electron gas also gives rise to such coupling between magnetic moments. This will be evaluated and compared with the effect of the polarisation of the valence band due to virtual interband transitions^{8), 9)}.

2. Polarisability of the non degenerate electron gas

We consider a system of magnetic ions or nuclei at positions \mathbf{R}_j and with spins \mathbf{S}_j , which act on an electron with coordinate \mathbf{r} and spin \mathbf{s} as an external potential

$$\sum_j -2 J(\mathbf{x} - \mathbf{R}_j) (\mathbf{s} \cdot \mathbf{S}_j). \quad (1)$$

Using Bloch waves

$$\varphi_{\mathbf{k}\sigma} = V^{-1/2} e^{i\mathbf{k}\mathbf{x}} u_{\mathbf{k}}(\mathbf{x}) \cdot \eta_{\sigma} \quad (2)$$

we define

$$J(\mathbf{k}, \mathbf{k}') = \Omega^{-1} \int e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{x}} u_{\mathbf{k}'}^*(\mathbf{x}) u_{\mathbf{k}}(\mathbf{x}) J(\mathbf{x}) d\mathbf{x} \tag{3}$$

where Ω is the volume of the atomic cell. In particular for a point interaction

$$J(\mathbf{x}) = J_0 \Omega \delta(\mathbf{x}) \tag{4}$$

(3) becomes

$$J(\mathbf{k}, \mathbf{k}') = J_0 u_{\mathbf{k}'}^*(0) u_{\mathbf{k}}(0). \tag{5}$$

With a development of the electron field in Bloch waves the interaction energy takes the form^{10),4)}

$$H' = - N^{-1} \sum_{\mathbf{k}'} \sum_{\mathbf{k}} \sum_j J(\mathbf{k}, \mathbf{k}') e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_j} \left\{ \begin{aligned} &(a_{\mathbf{k}'_+}^\dagger a_{\mathbf{k}_+} - a_{\mathbf{k}'_-}^\dagger a_{\mathbf{k}_-}) S_j^z + \\ &a_{\mathbf{k}'_+}^\dagger a_{\mathbf{k}_-} S_j^- + a_{\mathbf{k}'_-}^\dagger a_{\mathbf{k}_+} S_j^+ \end{aligned} \right\} \tag{6}$$

where $N = V/\Omega$ is the number of atoms. $a_{\mathbf{k}_\pm}^\dagger$ and $a_{\mathbf{k}_\pm}$ are creation and destruction operators for electrons with wave-vector \mathbf{k} and spin + or -.

The polarisation $P = n_+ - n_-$, where n_\pm is the density of electrons with + and - spin respectively, is obtained by perturbation theory to second order⁴⁾

$$P(\mathbf{x}) = \sum_j P_j(\mathbf{x}) \tag{7}$$

$$P_j(\mathbf{x}) = - \frac{2 S_j^z}{N^2 \Omega} \sum_{\mathbf{k}, \mathbf{k}'} u_{\mathbf{k}}^*(\mathbf{x}) u_{\mathbf{k}'}(\mathbf{x}) \frac{J(\mathbf{k}, \mathbf{k}')}{E(\mathbf{k}) - E(\mathbf{k}')} \left. \begin{aligned} &\cdot e^{-i(\mathbf{k}-\mathbf{k}')\cdot(\mathbf{x}-\mathbf{R}_j)} \cdot f(\mathbf{k}) \{1 - f(\mathbf{k}')\} + c. c. \end{aligned} \right\} \tag{8}$$

where $f(\mathbf{k})$ is the Fermi-Dirac distribution function.

The interaction energy up to second order is^{4),10)}

$$E' = \sum_i \sum_j A_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j) \tag{9}$$

$$A_{ij} = \frac{2}{N^2} \sum_{\mathbf{k}, \mathbf{k}'} \frac{|J(\mathbf{k}, \mathbf{k}')|^2}{E(\mathbf{k}) - E(\mathbf{k}')} e^{i(\mathbf{k}-\mathbf{k}')\cdot(\mathbf{R}_i-\mathbf{R}_j)} f(\mathbf{k}) \{1 - f(\mathbf{k}')\}. \tag{10}$$

$P_j(\mathbf{R}_i)$ and A_{ij} are related to each other. Let $\overline{J u^2}$ represent a weighted average of $J(\mathbf{k}, \mathbf{k}') u_{\mathbf{k}}^*(0) u_{\mathbf{k}'}(0)$ in (8) and $\overline{J^2}$ a weighted average of $|J(\mathbf{k}, \mathbf{k}')|^2$ in (10), then

$$A_{ij} / \overline{J^2} \Omega = - P_j(\mathbf{R}_i) / 2 \overline{J u^2} S_j^z. \tag{11}$$

It should be noted that $P(\mathbf{x})$ does not imply a variation of the total charge density, since the excess of plus spin electrons equals at every point the deficiency of minus spin electrons. Therefore correlation effects will not appear in this approximation. If, however, spin independent interactions are considered, correlation effects would have to be included¹¹⁾.

3. Evaluation of the integrals

We suppose that the energy is of the form

$$E(\mathbf{k}) = \frac{\hbar^2}{2} \left(\frac{k_x^2}{m_x} + \frac{k_y^2}{m_y} + \frac{k_z^2}{m_z} \right) \quad (12)$$

and introduce the variables

$$\kappa_x = k_x m_x^{-1/2}, \dots, \dots, \varrho_x = R_x m_x^{+1/2}, \dots, \dots \quad (13)$$

Since terms in $f(\mathbf{k}) f(\mathbf{k}')$ cancel, (10) becomes

$$A = \frac{4 \bar{J}^2}{N^2 \hbar^2} \sum_{\kappa, \kappa'} \frac{e^{i(\kappa - \kappa') \cdot \varrho}}{\kappa^2 - \kappa'^2} f(\kappa) \quad (14)$$

where we have dropped the index ij in A_{ij} and $\varrho_{ij} = \varrho_i - \varrho_j$. With polar coordinates the integration over the angles gives

$$A = \frac{\bar{J}^2 \Omega^2 m_x m_y m_z}{\pi^4 \hbar^2 \varrho^2} \int_0^\infty d\kappa \int_0^\infty d\kappa' \kappa \kappa' \frac{\sin(\kappa \varrho) \sin(\kappa' \varrho)}{\kappa^2 - \kappa'^2} f(\kappa). \quad (15)$$

After integration over κ' by principal parts

$$A(\varrho, \eta) = - \frac{\bar{J}^2 \Omega^2 m_x m_y m_z k_B T}{4 \pi^3 \hbar^4 \varrho^2} G(\varrho, \eta) \quad (16)$$

$$G(\varrho, \eta) = \frac{\hbar^2}{k_B T} \int_0^\infty d\kappa \kappa \sin(2 \kappa \varrho) f(\kappa). \quad (17)$$

Here

$$f(\kappa) = \left[e^{\frac{\hbar^2 \kappa^2}{2k_B T} - \eta} + 1 \right]^{-1} \quad (18)$$

so that (17) becomes

$$G(\alpha, \eta) = \int_0^\infty dx \sin(\alpha x^{1/2}) [e^{x-\eta} + 1]^{-1} \quad (19)$$

where

$$\alpha = 2 (2 k_B T)^{1/2} \hbar^{-1} \varrho. \quad (20)$$

This integral has been tabulated as a function of α for some values of η using the electronic computer of the ETH.

In the following limiting cases the integral is elementary. For a *degenerate gas*, $\eta \rightarrow \infty$, the formula of RUDERMAN and KITTEL is obtained

$$A(\varrho, \infty) = - \frac{\bar{J}^2 \Omega^2 m_x m_y m_z}{4 \pi^3 \hbar^2 \varrho^2} \int_0^{\kappa_F} d\kappa \kappa \sin(2 \kappa \varrho) \quad (21)$$

$$A(\varrho, \infty) = -\frac{\bar{J}^2 \Omega^2 m_x m_y m_z}{16 \pi^3 \hbar^2 \varrho^4} [\sin(2 \kappa_F \varrho) - 2 \kappa_F \varrho \cos(2 \kappa_F \varrho)] \quad (22)$$

where $\hbar^2 \kappa_F^2/2$ is the top of the energy surface. In the other extreme for the Boltzmann gas, $\eta \rightarrow -\infty$,

$$f(\kappa) = e^\eta e^{-\hbar^2 \kappa^2/2k_B T} \quad (23)$$

$$A(\varrho, -\infty) = -\frac{\bar{J}^2 \Omega^2 m_x m_y m_z}{4 \pi^3 \hbar^2 \varrho^2} e^\eta \int_0^\infty d\kappa \kappa \sin(2 \kappa \varrho) e^{-\hbar^2 \kappa^2/2k_B T} \quad (24)$$

$$= -\frac{\bar{J}^2 \Omega (m_x m_y m_z)^{1/2} n}{2 \pi \hbar} \frac{1}{\varrho} e^{-2k_B T \varrho^2/\hbar^2}. \quad (25)$$

The density of particles n is related to η through

$$n = \frac{(m_x m_y m_z)^{1/2}}{2 \pi^2} \left(\frac{2 k_B T}{\hbar^2} \right)^{3/2} F_{1/2}(\eta) \quad (26)$$

where $F_{1/2}$ is a tabulated²⁾ function. In particular

$$F_{1/2}(\eta \rightarrow -\infty) = \frac{\sqrt{\pi}}{2} e^\eta. \quad (27)$$

4. Discussion of the results

The RUDERMAN, KITTEL function (22) oscillates and diminishes with distance. The interaction between two magnetic moments in a degenerate electron gas is therefore alternately ferromagnetic and antiferromagnetic. The corresponding function (25) for the Boltzmann case falls off as a Gaussian curve at large distances. The resulting interaction is ferromagnetic for all distances. It is weaker, however, with comparatively long range $(2 k_B T)^{-1/2} \hbar$. The oscillation of (22) comes from the sharp cut off of the integration, which is replaced by a gradual decrease of the integrand with κ in (24).

As examples of *intermediate cases* Figure 1 shows $G(\varrho, \eta)$ for four values of η . Detailed tables of this integral will be published in a forthcoming paper. It can be seen that the curve for $\eta = 4$ resembles the Ruderman, Kittel function, whereas for $\eta = -2$ the behaviour of the Boltzmann case is obtained. Note that $G(\varrho, \infty)$ plotted in the units of Figure 1 would have vanishing range and $G(\varrho, -\infty)$ vanishing amplitude.

The interaction A is given as a function of

$$\varrho = (m_x R_x^2 + m_y R_y^2 + m_z R_z^2)^{1/2} \quad (28)$$

Hence the surfaces of equal interaction energy in real space are ellipsoids, the long axes pointing into the direction of small effective mass.

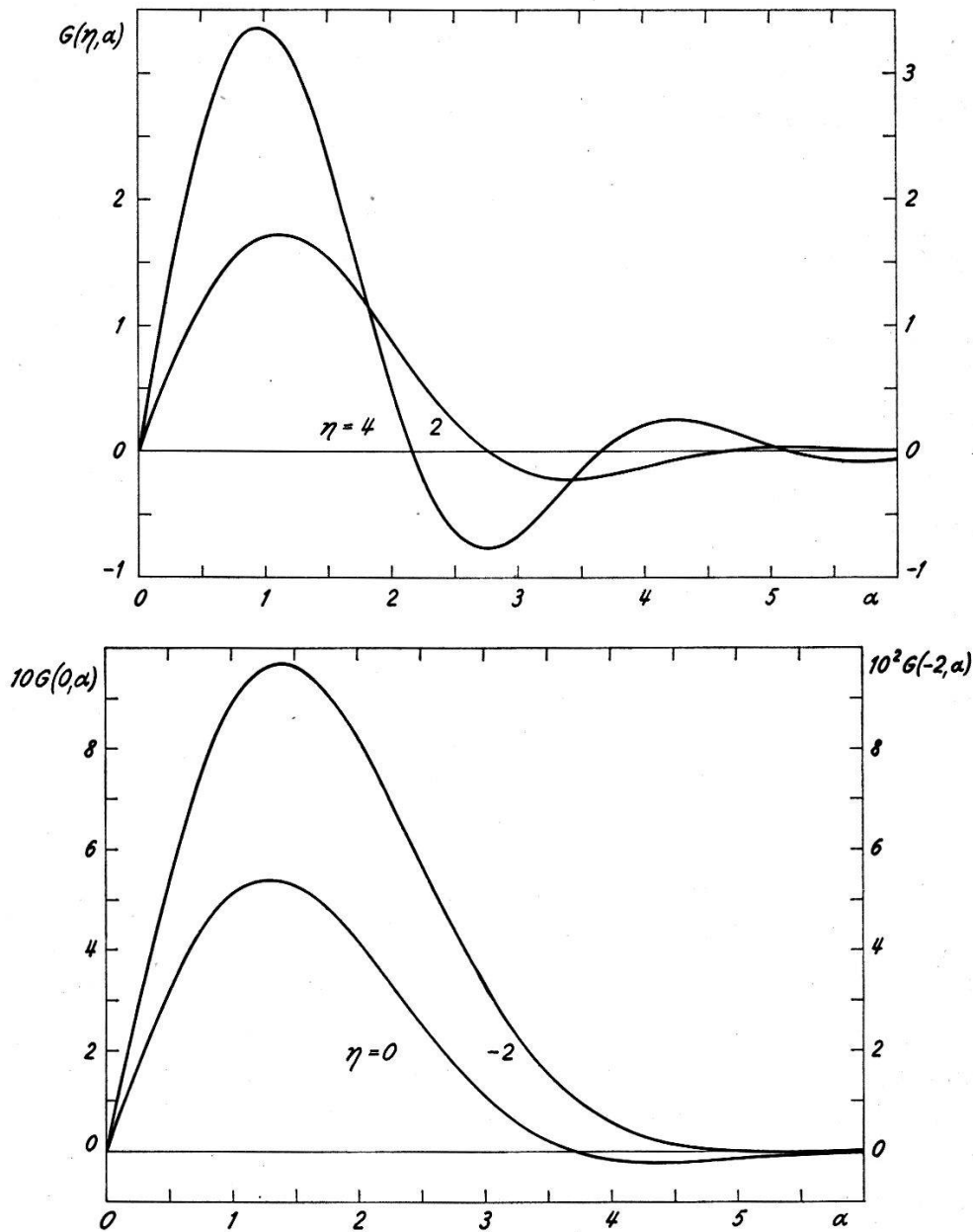


Fig. 1

Distance dependence of ρ^2 times the interaction between magnetic moments in an electron gas for some values of the degeneracy η

The polarisation due to a point source can be related to that produced by a homogeneous field. The energy of an electron in the field F is given by $-F s_z$ (for a magnetic induction B_z we must take $F = g \mu_B B_z$, where g is the spectroscopic splitting factor and μ_B the negative Bohr magneton). F can be written as a superposition of point fields

$$F = \frac{F}{(-2 J_0 \Omega S^z)} \int (-2 J_0 \Omega S^z) \delta(\mathbf{x}) d\mathbf{x}. \tag{29}$$

Since the polarisation $P_j(\mathbf{x}, \eta)$ caused by a point field at position \mathbf{R}_j is proportional to J_0 via $J(\mathbf{k}, \mathbf{k}') = J_0 u_{\mathbf{k}'}(\mathbf{R}_j) u_{\mathbf{k}}(\mathbf{R}_j)$ the polarisation due

to a homogeneous field can be expressed as an integral over point field polarisations:

$$P(\mathbf{x}, \eta) = \frac{F}{2 J_0 \Omega S_j^z} \int P_j(\mathbf{x}, \eta) d\mathbf{R}_j. \quad (30)$$

If we assume that $u_{\mathbf{k}}(\mathbf{x})$ is independent of \mathbf{k} and use equations (11), (13) and (16) this reduces to

$$P(\mathbf{x}, \eta) = \frac{F(m_x m_y m_z)^{1/2} \overline{u^2(\mathbf{x})} k_B T}{\pi^2 \hbar^4} \int_0^\infty G(\varrho, \eta) d\varrho. \quad (31)$$

$\overline{u^2}$ actually is a function of \mathbf{x} indicating the variation of $P(\eta)$ within each cell. For the Ruderman, Kittel case (31) reduces to the Pauli spin paramagnetism

$$P(\infty) = F \overline{u^2} \nu \quad (32)$$

where

$$\nu = \kappa_F (m_x m_y m_z)^{1/2} / 2 \pi^2 \hbar^2 \quad (33)$$

is the level density per unit volume at the Fermi surface. In the Boltzmann case

$$P(-\infty) = F \overline{u^2} n / 2 k_B T \quad (34)$$

the expression for the Curie law is obtained.

5. Interactions by virtual excitations from the valence band

The interaction between magnetic moments via a non degenerate electron gas may have an influence in semiconductors. Since the conduction electrons form a dilute gas of particles with low kinetic energy, the interaction is weak but of long range, whereas the valence electrons are capable of producing strong short range interactions.

This second interaction may be pictured as arising from the polarisation of the system of valence electrons. It is included in the expressions (8) and (10) if we allow \mathbf{k} and \mathbf{k}' to run through both the valence and conduction bands. $u_{\mathbf{k}}$ is a different function for the two bands. We shall consider the contribution coming from a full valence and an empty conduction band.

An evaluation of the integrals for a realistic case is not elementary. Models have been proposed^{8),9)} in which

$$-E(\mathbf{k}) + E(\mathbf{k}') = E_g + \frac{\hbar^2}{2 m_h} k^2 + \frac{\hbar^2}{2 m_e} k'^2 \quad (35)$$

where E_g is the direct optical energy gap and m_h and m_e are respectively the effective masses of valence and conduction electrons. Note that if the summation is extended to infinity for both \mathbf{k} and \mathbf{k}' , the resulting integral does not exist, unless special prescriptions for the integration are followed.

BLOEMBERGEN and ROWLAND⁸) assume the valence band to be narrow compared to E_g and neglect $(\hbar^2/2 m_h) k^2$ in (35). The integration over k is cut off at k_t , where

$$k_t = 2 \pi (3/4 \pi \Omega)^{1/3} \quad (36)$$

while that over \mathbf{k}' goes from 0 to ∞ to include contributions from higher bands. They obtain an interaction of the form (9) where

$$A_{ij} = - \frac{\bar{J}^2 \Omega^2 m_e}{2 \pi^3 \hbar^2 R_{ij}^4} [\sin(k_t R_{ij}) - k_t R_{ij} \cos(k_t R_{ij})] e^{-\frac{(2m_e E_g)^{1/2}}{\hbar} R_{ij}}. \quad (37)$$

The polarisation (8) now is a sum over the functions $v_{\mathbf{k}}(\mathbf{x}) u_{\mathbf{k}'}(\mathbf{x}) e^{-i(\mathbf{k}-\mathbf{k}')\mathbf{x}}$, where $v_{\mathbf{k}} e^{i\mathbf{k}\mathbf{x}}$ and $u_{\mathbf{k}'} e^{i\mathbf{k}'\mathbf{x}}$ are Bloch functions of the valence and conduction band respectively. Since these Bloch functions are all orthogonal (even for $\mathbf{k} = \mathbf{k}'$) the polarisation assumes positive and negative values in each cell. Therefore the polarisation for a homogeneous field (30) vanishes. This also follows directly from the fact that a homogeneous field has vanishing matrix elements for interband-transitions. This means that there is no direct spin contribution to the van Vleck paramagnetism.

6. Numerical applications

For a dilute system of magnetic impurities embedded in a semiconductor, the numerical values of the various interactions are estimated in Table I for a semiconductor like germanium. We take $m_x = m_y = m_z = m_h = m_e = 1/10 m$, $\Omega = 2.24 \cdot 10^{-23} \text{ cm}^3$, and $E_g = 0.8 \text{ eV}$. The parameters which still can be varied are the distance between two magnetic ions R , the temperature T and the degeneracy η or the density of con-

Table I

Numerical values for the various interactions: the interaction via conduction electrons A , the envelope to the interaction due to the valence band A' , and the magnetic dipole interaction estimated as $A'' = \mu_B^2/R^3$

R	10				40				\AA
	100		500		100		500		$^\circ\text{K}$
η	2	-1	2	-1	2	-1	2	-1	
n	$4.3 \cdot 10^{17}$	$5.0 \cdot 10^{16}$	$4.8 \cdot 10^{18}$	$5.6 \cdot 10^{17}$	$4.3 \cdot 10^{17}$	$5.0 \cdot 10^{16}$	$4.8 \cdot 10^{18}$	$5.6 \cdot 10^{17}$	cm^{-3}
A/\bar{J}^2	$4.3 \cdot 10^{-8}$	$5.1 \cdot 10^{-9}$	$4.2 \cdot 10^{-7}$	$5.2 \cdot 10^{-8}$	$6.4 \cdot 10^{-9}$	$8.8 \cdot 10^{-10}$	$1.2 \cdot 10^{-9}$	$2.0 \cdot 10^{-9}$	eV [J in eV]
$ A' /\bar{J}^2$	$3.5 \cdot 10^{-5}$				$7.1 \cdot 10^{-9}$				eV [J in eV]
A''	$5.4 \cdot 10^{-8}$				$8.4 \cdot 10^{-10}$				eV

duction electrons n . The exchange integral J between a magnetic inner shell and a conduction electron may be a few tenths of an electron volt.

Table I shows that for $R = 10 \text{ \AA}$ the interaction via the valence band dominates, whereas for 40 \AA the various interactions considered may be of comparable magnitude. It may be noted that A increases with the density of the conduction electrons and with their effective mass. To obtain the density of conduction electrons used in Table I doping is necessary.

In a paramagnetic resonance experiment, the interactions give rise to a line width. For one Bohr magneton the energy of $6.7 \cdot 10^{-8} \text{ eV}$ corresponds to a width of one Oersted. The actual line shape involves the simultaneous interaction of many magnetic moments, a problem which has not been treated here.

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