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Indirect exchange interaction in parabolic symmetry-induced zero gap semiconductors

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Résumé. — L'interaction d'échange indirecte entre spins localisés est calculée en tenant compte des transitions virtuelles entre bande de valence et bande de conduction des semiconducteurs de gap nul. Elle décroît comme R^{-4} où R est la distance entre deux spins et est antiferromagnétique pour les matériaux existants. Le signe de l'interaction est imposé par la structure de bande. La température de Curie-Weiss des alliages $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ de faible x est en bon accord avec l'expérience.

Abstract. — The indirect exchange interaction between two localized spins is calculated, taking into account the virtual interband transitions across the zero gap. This interaction decreases like R^{-4} , where R is the distance between two spins, and is antiferromagnetic for all existing materials. The sign of the interaction crucially depends on the band structure. The calculated Curie-Weiss temperature of $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ alloys with low x is in good agreement with the experiments.

1. Introduction. — Recent experiments on the symmetry-induced zero gap $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ alloys have evidenced an antiferromagnetic interaction between localized Mn^{2+} moments ($L = 0$, $S = J = 5/2$, $g_{\text{Mn}} = 2$): high temperature susceptibility measurements [1, 2] revealed a Curie-Weiss behaviour $\chi^{-1}(T) = a(T + \theta)$ $\theta > 0$, whereas low temperature interband magneto-absorption [3] as well as magneto-transport phenomena [4] evidence a significant antiferromagnetic interaction between localized spins. Neither of these experiments could be successfully explained by only taking into account statistical distributions of isolated, paired, tripled... Mn^{2+} moments, i.e. assuming short range interactions. The purpose of the present letter is to present a model of long range indirect exchange interaction, which takes care of the virtual electronic transitions across the zero gap between the filled valence band and the almost empty conduction band. The letter will be organized as follows: in section 2 we derive the eigenfunctions of the effective Luttinger Hamiltonian [5] describing Γ_8 bands. In section 3, we calculate the effective interaction between the localized moments. In section 4, we compare the Curie-Weiss

temperature θ taken from our model with the experimental results.

2. Wavefunctions. — The symmetry-induced zero gap $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ alloys have a band structure which is inverted with respect to InSb-like materials (Fig. 1). The conduction and valence bands have the same Γ_8 symmetry at the centre of the Brillouin zone. The valence band has a heavy mass ($m_v \approx 0.4 m_0$), whereas the conduction band has a small effective

Fig. 1. — Band structure of symmetry-induced zero gap semiconductors.

mass ($m_c \approx 3 \times 10^{-2} m_0$ in HgTe) which decreases almost proportionally to $|\varepsilon_0|$. In spite of non parabolic phenomena, which are crucial for alloys with small $|\varepsilon_0|$, we shall restrict our considerations to parabolic zero gap materials, i.e. alloys with $x \lesssim 1\%$.

For such small x values the experimental Curie-Weiss temperature $\theta_{\text{exp}} \lesssim 6$ K. Then the Curie-Weiss behaviour $\chi^{-1} = a(T + \theta)$ is easily observed for $15 \text{ K} < T < 50 \text{ K}$. At these low temperatures there are practically no free carriers ($n, p < 10^{16} \text{ cm}^{-3}$) [6], and we will be dealing with the genuine interband $\Gamma_8^v \rightarrow \Gamma_8^c$ effect. Consequently, we will assume $k_F = 0$: a filled valence band and an empty conduction band.

For parabolic Γ_8 bands, the effective Luttinger Hamiltonian [5] corresponding to $J = 3/2$ spinors can be written :

$$\mathcal{H}(k) = \frac{1}{8} \left[\frac{9}{2} \frac{\hbar^2}{m_c} + \frac{1}{2} \frac{\hbar^2}{m_v} \right] k^2 - \frac{1}{2} \left[\frac{\hbar^2}{2 m_v} + \frac{\hbar^2}{2 m_c} \right] (\mathbf{k} \cdot \mathbf{J})^2. \quad (1)$$

When projected on the basis

$$|3/2, 3/2\rangle; |3/2, -1/2\rangle; |3/2, 1/2\rangle; |3/2, -3/2\rangle,$$

the eigenstates corresponding to positive eigenvalues $\varepsilon_c(k) = \hbar^2 k^2 / 2 m_c$ are ($\mathbf{k} = k, \theta, \varphi$) :

$$\psi_{e+} = \frac{\exp i\mathbf{k} \cdot \mathbf{r}}{\sqrt{\Omega}} \begin{bmatrix} -\frac{\sqrt{3}}{2} \sin \theta e^{-i\varphi} \\ \frac{1}{2} \sin \theta e^{i\varphi} \\ \cos \theta \\ 0 \end{bmatrix} \quad \psi_{e-} = \frac{\exp i\mathbf{k} \cdot \mathbf{r}}{\sqrt{\Omega}} \begin{bmatrix} 0 \\ \cos \theta e^{-i\varphi} \\ -\frac{1}{2} \sin \theta e^{-2i\varphi} \\ \frac{1}{2} \sqrt{3} \sin \theta \end{bmatrix} \quad (2)$$

whereas the wave functions of valence band electrons corresponding to $\varepsilon_v(k) = -\hbar^2 k^2 / 2 m_v$ are :

$$\psi_{h+} = \frac{\exp i\mathbf{k} \cdot \mathbf{r}}{\sqrt{\Omega}} \begin{bmatrix} \cos \theta e^{-3i\varphi} \\ 0 \\ \frac{\sqrt{3}}{2} \sin \theta e^{-2i\varphi} \\ \frac{1}{2} \sin \theta \end{bmatrix} \quad \psi_{h-} = \frac{\exp i\mathbf{k} \cdot \mathbf{r}}{\sqrt{\Omega}} \begin{bmatrix} -\frac{1}{2} \sin \theta e^{-3i\varphi} \\ -\frac{\sqrt{3}}{2} \sin \theta e^{-i\varphi} \\ 0 \\ \cos \theta \end{bmatrix}. \quad (3)$$

Note in eqs. (2), (3) that, in contrast to simple isotropic metals, the periodic parts of the Γ_8 Bloch functions explicitly depend on the direction of $\mathbf{k}$.

3. Indirect exchange interaction. — The mobile (in Γ_8 bands) and localized electrons (which form the magnetic moments) are assumed to interact *via* the Heisenberg type Hamiltonian :

$$\mathcal{H}_{\text{int}} = \sum_{\mathbf{R}_i} J(\mathbf{r} - \mathbf{R}_i) \mathbf{S}_i \cdot \boldsymbol{\sigma} \quad (4)$$

where $\mathbf{S}_i$ are localized spins located at $\mathbf{R}_i$ and $\boldsymbol{\sigma}$ the bare electron spin. $J(\mathbf{r} - \mathbf{R}_i)$ is an exchange integral centred at $\mathbf{R}_i$ and varying rapidly over a unit cell.

The first order energy shift produced by $\mathcal{H}_{\text{int}}$

vanishes, since it is proportional to the localized spin magnetization, which we assume to be zero (paramagnetic state). The second order correction is :

$$\Delta \varepsilon^{(2)} = \sum_{\substack{i \neq j \\ \mathbf{k} \neq \mathbf{k}'}} \frac{f(\varepsilon_{ik}) [1 - f(\varepsilon_{jk'})]}{\varepsilon_{ik} - \varepsilon_{jk'}} |\langle i\mathbf{k} | \mathcal{H}_{\text{int}} | j\mathbf{k}' \rangle|^2 \quad (5)$$

which, owing to the rotational invariance of the unperturbed Hamiltonian, may be rewritten in the form of an effective spin-spin Hamiltonian :

$$\Delta \varepsilon^{(2)} = \sum_{i > j} I(\mathbf{R}_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j. \quad (6)$$

Owing to the lack of free carriers $I(\mathbf{R}_{ij})$ consists only of an interband part

$$I_{ij}^{\text{inter}} = \frac{2}{\Omega^2} \sum_{\mathbf{k}, \mathbf{k}'} \frac{f[\varepsilon_{v\mathbf{k}'}] [1 - f(\varepsilon_{c\mathbf{k}})]}{\varepsilon_{v\mathbf{k}'} - \varepsilon_{c\mathbf{k}}} \exp i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}_{ij} \sum_{\sigma, \sigma' = \pm} |\langle u_{c\mathbf{k}\sigma} | J(r) \sigma_z | u_{v\mathbf{k}'\sigma'} \rangle|^2 \quad (7)$$

where the rotational invariance has again been used to express all the required matrix elements in terms of those involving σ_z .

To calculate I_{ij}^{inter} , we have to evaluate the matrix elements of $J(\mathbf{r})\sigma_z$ between the $k \neq 0$ and $k' \neq 0$ valence and conduction Bloch functions accounting

for the Kramers degeneracy of each band. The presence of a large spin orbit coupling leads to non vanishing matrix elements between states of Kramers doublets. Denoting by β the matrix $\langle x | J(\mathbf{r}) | x \rangle$, replacing σ_z by $\frac{1}{3}J_z$ (valid in the Γ_8 multiplet), and expressing wave vectors in spherical coordinates, one obtains :

$$\sum_{\sigma\sigma'=\pm} |\langle \mathbf{ck}\sigma | J(\mathbf{r})\sigma_z | \mathbf{vk}'\sigma' \rangle|^2 = \beta^2 \left\{ -\frac{5}{16} \cos^2 \theta \cos^2 \theta' + \frac{5}{48} - \frac{\cos^2 \theta}{16} + \frac{13}{48} \cos^2 \theta' - \frac{1}{16} \sin 2\theta \sin 2\theta' \cos(\varphi - \varphi') + \frac{1}{4} \sin^2 \theta \sin^2 \theta' \cos 2(\varphi - \varphi') \right\}. \quad (8)$$

The terms involving $(\varphi - \varphi')$ will vanish owing to the cylindrical symmetry around the R_{ij} axis. After straightforward and tedious calculations, we find :

$$I_{ij}^{\text{inter}}(k_F = 0) = -\frac{\beta^2 m_c m_v}{4 \Pi^3 \hbar^2 R^4} \int_0^\infty x^2 dx \int_0^\Pi \sin \theta d\theta e^{-ix \cos \theta} \int_0^\infty \frac{t^2 dt}{m_c t^2 + m_v x^2} \int_0^\Pi \sin \theta' d\theta' \times \\ \times e^{it \cos \theta'} \left\{ -\frac{5}{16} \cos^2 \theta \cos^2 \theta' + \frac{5}{48} - \frac{1}{16} \cos^2 \theta + \frac{13}{48} \cos^2 \theta' \right\} \quad (9)$$

$$I_{ij}^{\text{inter}}(k_F = 0) = -\frac{\beta^2 m_v}{16 \Pi^3 \hbar^2 R^4} f(r) \quad (10)$$

where

$$f(r) = -\frac{r}{1+r^2} + \frac{\Pi}{2} - \text{Arctg } r + \frac{13}{3} \frac{1}{r} - \frac{13}{3} \frac{1}{r^2} \text{Arctg } r \quad (11)$$

where

$$r = \sqrt{\frac{m_v}{m_c}} \quad \text{and} \quad R = |\mathbf{R}_{ij}|.$$

The variation of $f(r)$ upon r is shown on figure 2. The symmetry-induced zero gap semiconductors display an indirect exchange mechanism whose *sign* depends on the ratio between the valence and conduction masses. $f(r)$ shows a sign reversal near $r \approx 0.5$, and behaves like $-\frac{13 \Pi}{2} r^{-2}$ at large r . This sign

Fig. 2. — Plot of $f(r)$ versus r .

reversal crucially depends upon the band structure. Its origin may be traced back to the matrix elements of $J(\mathbf{r})\sigma_z$, and more precisely to the $\cos^2 \theta$ and $\cos^2 \theta'$ terms appearing in eq. (8). The sign reversal is absent for angular independent matrix elements, i.e. in the case of accidental band degeneracy, one finds $f(r) \geq 0$. For all existing zero gap materials $r > 3$: the indirect exchange mechanism due to the virtual interband transitions is then antiferromagnetic.

I_{ij} falls at large distance like R^{-4} , this dependence being imposed by the dimensionality of the problem. The power law variation is due to the absence of an energy gap between the valence and conduction bands. In the case of a finite gap it has been shown [7], by using an oversimplified dispersion law and a constant matrix element, that $I_{ij}(R)$ decreases exponentially. However, for zinc blende open gap materials, a complicated $I_{ij}(R)$ originating from angular matrix elements has to be expected.

4. Discussion : the magnetic susceptibility of zero gap $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ alloys. — The indirect exchange interaction due to virtual interband transitions is not

weak : for a material with band parameters similar to HgTe : $R \approx 6.4 \text{ \AA}$ (second nearest neighbour), and $N_0 \beta = 1.5 \text{ eV}$ [3, 4] where N_0 is the number of unit cells per unit volume, one finds $I_{ij} \approx 0.2 \text{ meV}$. The interband mechanism provides a natural explanation of the observed antiferromagnetic interaction between Mn^{2+} localized moments already reported in the high temperature susceptibility measurements [1, 2]. In the molecular field approximation [8] (high T), one finds for dilute alloys

$$\chi^{-1}(T) = a(T + \theta) \quad \theta > 0 \quad (12)$$

where

$$k_B \theta = \frac{35}{12} x \frac{\beta^2 m_v |f(r)|}{16 \pi^3 \hbar^2 a_0^4} \sum_{j \neq 0} \left(\frac{a_0}{R_{j0}} \right)^4 \quad (13)$$

x is the Mn content and the summation runs over all the sites of an fcc lattice with lattice parameter a_0 . If we calculate θ for an alloy with $x = 1 \%$ and assume that the summation over lattice sites is of

the order of 94, we get $\theta \approx 6.9 \text{ K}$, in agreement with experiments [1, 2] ($\theta_{\text{exp}} \approx 6 \text{ K}$).

The main limitation of our model is the absence of any non parabolic effects in the Γ_8^c electron wave functions and dispersion relations. This is evidenced by looking at eqs. (10), (11), which predict $I_{ij} = 0$ if ε_0 and $m_c = 0$ ($r = \infty$). Such a result originates from the fact that a parabolic law is progressively less appropriate for describing the dispersion relations of alloys as ε_0 becomes smaller. A complete inaccuracy is even obtained in the case of $\varepsilon_0 = 0$ for which $\varepsilon_c(k) = Ak$. Our treatment is then clearly limited to parabolic alloys $|\varepsilon_0| > 200 \text{ meV}$, $x < 1 \%$. More detailed results concerning the non parabolic zero gap alloys will be published elsewhere.

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