INTERLAYER COUPLING IN MAGNETIC SUPERLATTICES WITH ELECTRON DENSITY INHOMOGENEITIES

W. Gruhn, Z. Bąk
Institute of Physics, Pedagogical University,
al. Amrii Krajowej 13/15, 42-200 Częstochowa

ABSTRACT

We discuss the influence of spatial inhomogeneities of the free electron density on the magnetic interaction between magnetic layers of the superlattice, mediated across nonmagnetic, metallic spacer. Using the modified total energy approach, we prove that for TM or RE superlattices the additional scattering of free electrons on magnetic ion multipole moment increases the ferroquadrupolar biquadratic coupling between magnetic layers. We show that this novel mechanism, should manifest itself in the anisotropy of the magnetoresistivity.

INTRODUCTION

The indirect magnetic coupling between ferromagnetic (FM) layers across a nonmagnetic (NM) metal spacer has been extensively studied both experimentally and theoretically. An oscillatory dependence of the magnetic coupling strength on the thickness of the spacer layer has been observed in a large number of systems (metallic magnetic superlattices) [1,2,3,4]. According to the theory of RKKY-like coupling [5], the problem of magnetic interlayer exchange coupling includes two aspects: firstly, the interaction between ferromagnetic layer and conduction electrons of nonmagnetic metal layers; secondly, the way of the spin polarization propagating through the nonmagnetic metal spacer.

Fabrication of strained layers offers a high potential for the design of new multiple quantum wells. Apart from the critical thickness, the stress is relaxed by *e.g.* misfit dislocations. This in turn generates lateral strain

modulation and interface undulations observed in many heterostructures (see [6,7] and references therein). In this paper we will focus our consideration on the case of magnetic, metallic superlattices (MSL) and we will study the effect of lateral inhomogeneities on the magnetic coupling between magnetic layers across metallic, nonmagnetic spacer. In metallic systems, magnetic interactions are propagated by itinerant electrons and thus can be transmitted over relatively long distances. As a result of this, magnetic layers become coupled through nonmagnetic metallic layers. Most theoretical studies of the interlayer coupling in the MSL system limits to the exchange interaction being bilinear in spin or in-layer magnetization. However, in some MSL system biquadratic [8,9] or generally quadrupolar interactions [10] need to be considered. Almost all theoretical studies of the problem are based on restrictive assumption on the system: the layers are supposed to be coherent, *i.e.* it is assumed that the in-plane translational symmetry is not broken. This is true for those heteroepitaxial MSL for which the elemental consituents exhibit negligible ionic radii mismatch. When the ionic radii differ by a few percent, in the interface region arises additional tensile or compressive strains relaxed through misfit dislocations. The indirect magnetic coupling between neighbouring magnetic layer across the nonmagnetic spacer layer depends on the density of itinerant electrons which in the case of interface undulations (generated due to nonuniform lateral strains) also becomes non-uniform [11]. The aim of the paper is to study the effect of lateral strain undulations on the biquadratic interlayer exchange that arises from direct ionic-quadrupole and free electron spin scattering [10].

The effect of nonuniform interface undulation on the free electron density, in the case of sinusoidal modulation of the misfit stress has been calculated in [11]. The in-plane variation of lattice distortion arising because of this stress is the source of additional (compared to state of nondistorted lattice) potential experienced by the free electrons. The

resulting density of free electron states within the spacer layer is given by the equation [11,12]:

$$\rho(r) = \rho_0 \cdot [1 - p \cdot \cos(\vec{Q} \cdot \vec{r})] \qquad (1)$$

where ρ_0 – uniform free electron density,

p – amplitude of modulation,

Q – wave-vector of the planar superstructure.

This lateral density modulation, superposed with the charge density variation associated with the layered structure, results in multi-Q modulation of the free electron density of the form:

$$\rho = \rho_0 \cdot [1 + \sum_{q_0} \eta_{q_0} \cdot e^{iq_0 \cdot r}] \cdot \qquad (2)$$

In the theoretical description of the interlayer coupling, two alternative approaches can be applied. We can use either the modified RKKY approach [5] or the total energy calculation method [14,15]. The latter, based on the picture of free electrons confined within quantum well is the most general and transparent. However, till now all the calculations of intelayer coupling assumed the uniform free electron density. In the following, within total energy approach, we will derive the formula for strength of interlayer coupling with non-uniform free electron density taken in to account. We will assume that misfit strain is the main source of inhomogeneity and we will focus our considerations on the biquadratic interaction. Usually in the description of magnetic superlattices it is assumed that:

i) the in-layer translational symmetry is not broken,

ii) the electronic configurations of the magnetic ions are stable.

Experimental data show that in many superlattices either one or both assumptions are not fulfilled.

Since the free electron density within the spacer layer is given by the charge-density wave solution, it is evident that itinerant electron wave-function are no longer given by the free-electron planewaves but by the

CDW eigenfunctions [12]. In the simplest case of single-Q modulated conduction electron density, the pure single-Q CDW state is given by the following equation [12,13]:

$$\Phi_k\left(\vec{Q},\vec{r}\right) \approx e^{i\vec{k}\cdot\vec{r}} + \alpha_+ e^{i\left(\vec{k}+\vec{Q}\right)\cdot\vec{r}} + \alpha_- e^{i\left(\vec{k}-\vec{Q}\right)\cdot\vec{r}} \qquad (3)$$

where α is the amplitude of oscillation.

Determination of the eigenfunctions of mobile electrons (2) (within the spacer layer) let us calculate the modification of the interlayer coupling generated due to the interface undulations. We will apply the modified quantum well electron confinement approach of Bruno [14]. Application of this method to the conventional RKKY problem of magnetic impurities in a host metal, when the coupling of the ionic spin S_n with the itinerant electron spin σ is usually taken as the contact interaction

$$H_{ex} = -2JS_n \cdot \sigma\delta(r) \qquad (4)$$

leads to bilinear in layer magnetizations, RKKY-reminiscent interlayer coupling. There are minor modifications of the coupling constants, arising from the asphericity of the Fermi surface. However, if we consider the case of quadrupolar coupling generated by the contact electron spin-ionic quadrupole scattering [10,18,19]

$$V_{qc} = \frac{D}{k_F^2} \sum_{k^+k^-\sigma^+\sigma^-} \left[\left(\vec{S}_n \cdot \vec{k}^+\right)\left(\vec{S}_n \cdot \vec{k}^-\right) - \frac{1}{3}S(S+1)\vec{k}^+ \cdot \vec{k}^- \right] c^+_{k^+\sigma^+} c_{k^-\sigma^-} \qquad (5)$$

the effect of interface strain becomes more important. In equation (5) k^+, σ^+, k^-, σ^- denote the spin and wave-vector of the impact and the scattered electron, respectively. D is a constant that determines the strength of the scattering potential.

Using the scattering potential (5), within perturbative approach, we have shown, that the interaction between quadrupole moments of magnetic ions (Q_i and Q_j) is given by the following equation [10,20]:

$$H(R_{ij}) = -\sum_{\alpha\beta\gamma\delta} Q_{\alpha}^{\beta} \cdot Q_{\gamma}^{\delta} \cdot \Lambda_{\alpha\beta}^{\gamma\delta}\left(\left|R_i - R_j\right|\right), \qquad (6)$$

where the Q_{α}^{β} denotes the respective components of ionic quadrupole moment:

$$Q_{\alpha}^{\beta} = S_n^{\alpha} \cdot S_n^{\beta} - \frac{1}{3} S(S+1)\delta_{\alpha\beta}. \qquad (7)$$

Similarly to the case of bilinear spin-spin exchange, the interaction (5) favours the ferromagnetic (ferroquadrupolar) coupling [10]. However, the perturbative method suffers many shortcomings [20], first of all it gives interaction between individual ionic moments, while in the MSL we are interested in the entire coupling of adjacent magnetic layers. The latter coupling, however, can be determined within the quantum well confinement method [15].

The model system consists of a metallic spacer layer (paramagnetic), sandwiched between two potential perturbations of height V^A and V^B, generated by magnetic layers adjacent to the spacer. The effective coupling between magnetic sheets across the spacer is associated with the multiple internal reflections of free electrons at the interfaces. The resulting "quantum interference" produces interlayer coupling that oscillates with a period directly related to the geometry of the system. As it was shown by Bruno [15], the energy change due to the quantum interferences within spacer is given by the following equation:

$$\Delta E = \frac{2}{\pi}\int_{\infty}^{\varepsilon_F} \ln\left|1 - r_A r_B \cdot e^{2ik_{\perp}L}\right| d\varepsilon \approx \frac{1}{\pi^3}\int d^2k_{\parallel} \int_{-\infty}^{\varepsilon_F} r_A r_B \cdot e^{2ik_{\perp}L} \cdot d\varepsilon \qquad (8)$$

where r_A and r_B are the reflection amplitudes at both interfaces, L is the spacer thickness $L = (N+1)d$, with d being the thickness of one atomic plane in the spacer, while $k_{\perp}$ denotes the perpendicular to interface component of the impact electron wave-vector. We must note, however,

that contrary to the approach presented in [15,20] to the problem under consideration, the scattered electrons are described by the wavefunctions (3). This means that the impact and scattered electrons states are described by the wave-vectors $k^+ = \kappa^+$ and $k^+ = \kappa^+ \pm Q$ or $k^- = \kappa^-$ and $k^- = \kappa^- \pm Q$ respectively, where κ denotes the electron quasi-momentum, while Q is the superstructure modulation wave-vector.

Let us assume that the magnetizations of the ferromagnetic layers are at angle θ with respect to each other and determine the coupling energy ΔE (8) as the function of the θ angle. In the zeroth approximation, the reflection amplitude at the interface F_A is given by the following equation[14]:

$$r_A = A\left(k_\perp^+, k_\perp^-\right) \cdot \left\langle \vec{k}^- \middle| V^A \middle| \vec{k}^+ \right\rangle = A\left(k_\perp^+, k_\perp^-\right) \cdot \left\langle \vec{k}^- \middle| V_0^A + V_{qc}^A \middle| \vec{k}^+ \right\rangle = r_A^0 + r_A^{qc} \qquad (9)$$

where

$$r_A^0 = A\left(k_\perp^+, k_\perp^-\right) \cdot \left\langle \vec{k}^- \middle| V_0^A \middle| \vec{k}^+ \right\rangle = A(k_z) \left\langle \vec{k}^- \middle| V_0^A \middle| \vec{k}^+ \right\rangle \qquad (10)$$

defines the contribution from isotropic potential as well as from exchange contact interaction given by (4). r_A^{qc} is the reflection amplitude due to the electron-quadrupole scattering (5). Let us focus our attention on this term. In the equation (10) we should consider that wave-vectors of the incident $\vec{k}^+$ and reflected $\vec{k}^-$ electron fulfill the relation $\vec{k}_\perp^+ = -\vec{k}_\perp^-$. To take into account the effect of magnetization canting one must note that the scattering due to the quadrupolar potential (5) does not depend on the spin of impact electron. This means that the electron reflection amplitudes are equal, *i.e.* $r_{AB}^{qc\uparrow} = r_{AB}^{qc\downarrow}$. Thus we must modify the approach of Bruno [15], who exploits the spin dependence of the reflection amplitudes. To simplify our further considerations, we will replace the spin operators S_i in the Eq. (5) by their average, *i.e.* the layer magnetization M. Provided that the

parallel to interface component of the impact electron wave-vector forms angle ϕ with the in-plane magnetization of the F_A-th layer, in view of Eq. (5), we can write the reflection amplitudes as:

$$r_A^{qc}(\phi) \approx \left\langle \vec{k}^- \left| V_{qc}^A \right| \vec{k}^+ \right\rangle = C_3 \left[C_1 \cos^2 \phi + C_2^A \right] \qquad (11)$$

Similarly, considering the fact that magnetization of the second layer is parallel to the layer and forms angle θ with magnetization of the F_B-th layer, we can write

$$r_B^{qc}(\phi \pm \theta) \approx \left\langle \vec{k}^- \left| V_{qc}^B \right| \vec{k}^+ \right\rangle = C_3 \left[C_1 \cos^2(\phi \pm \theta) + C_2^B(\theta) \right] \quad (12)$$

where

$$C_1 = M^2 k_\parallel^2 \left(1 + 2\alpha^2\right), \qquad (13)$$

$$C_2^A = 2\alpha^2 Q^2 \cos^2 \delta - \frac{1}{3} S(S+1) \left[\left(1 + 2\alpha^2\right)\left(k_\parallel^2 - k^2\right) + 2\alpha^2 Q^2 \right], \quad (14a)$$

$$C_2^B = 2\alpha^2 Q^2 \cos^2(\delta \pm \theta) - \frac{1}{3} S(S+1) \left[\left(1 + 2\alpha^2\right)\left(k_\parallel^2 - k^2\right) + 2\alpha^2 Q^2 \right], \quad (14b)$$

$$C_3 = \frac{-a^2 m^2}{\hbar^4} \left(\frac{1}{1 + |\alpha|^2} \left[\frac{2m}{\hbar^2} \left(\varepsilon_k - 2G\alpha \right) - Q^2 |\alpha|^2 \right] - k_\parallel^2 \right)^{-1} \qquad (15)$$

Integration over $k_\parallel$ in Eq. (5) performed in polar coordinates $\left(k_\parallel, \phi\right)$ gives us

$$\int_0^{2\pi} d\phi \cdot r_A^{qc}(\phi) r_B^{qc}(\phi, \theta) \approx \int_0^{2\pi} C_3^2 \left[C_1 \cos^2 \phi + C_2^A \right] \left[C_1 \cos^2(\phi + \theta) + C_2^B \right]. \qquad (16)$$

Having the result (15), we can find that the energy change ΔE given by formula (8) depends on the θ as in the following equation:

$$\Delta E(\theta) = \Delta E(\cos^2\theta) \approx \Delta E^0 + J_2^Q \cos^2\theta + J_2^{DM} \cos\theta \sin\theta + \ldots , \quad (17)$$

where

$$J_2^Q = \pi \int dk_{\parallel} \int d\varepsilon C_3^2 \left[\frac{1}{2} C_1^2 + \left(C_1 + C_2^A \right) 2\alpha^2 Q^2 \left(2\cos^2\delta - 1 \right) \right] \quad (18)$$

$$J_2^{DM} = 2\pi \int dk_{\parallel} \int d\varepsilon C_3^2 \left(C_1 + C_2^A \right) 2\alpha^2 Q^2 \cos\delta \sin\delta \quad (19)$$

The coefficient J_2^Q, is the isotropic, biquadratic exchange integral, while second coefficient J_2^{DM} is the quartic Dzialoshynski-Moriya interaction [21,22,26]. The ratio $J_2^Q / J_2^{DM} \sim \alpha^2$ this means that the isotropic term dominates the DM interaction. Existence of such interactions was suggested by Rühriga *et al.* [23]. In the paper by Xia *et al.* [24] it was shown that bilinear Dzialoshynski-Moriya interaction can arise even in the absence of superlattice strains.

In view of the result (17), we can claim that we have proved within the total energy calculation method that the scattering potential (5) is the source of biquadratic exchange between magnetic layers. The interface undulations are a source of additional contribution to the biquadratic exchange integral J_2.

In summary, we have considered modifications if the interlayer coupling in the MSL systems arising from the lateral strains generated at the interfaces. Contrary to the other works considering the biquadratic coupling [25] we start from direct contact potential (5) and then apply the modified quantum confinement approach [20] to the multiple quantum well system [14]. The approach presented above gives a precise, quantitative relationship between the strength of the quadrupolar scattering potential and the biquadratic exchange integral J_2. In view of the general properties of the quantum interferences due to the electron

confinement within spacer, the effective biquadratic exchange integral oscillates with the spacer thickness [15]. In the above considerations, we have assumed that the interface lattice undulations are described by the wave-vector Q, which is parallel to the layers. However, in some MSL spacer undulations in the MSL, growth directions are observed [7]. Within the approach presented above this would mean that there should appear a second period of the magnetic oscillations being commensurate with the lattice undulation period.

Appendix

Electron spectrum within nonmagnetic interlayer in the case of lateral undulation can be determined as follows. The Hamiltonian of mobile electrons within the space can be written as [17,11]:

$$H(r) = -\frac{\hbar^2}{2m}\nabla^2 + V_0 + G\cos(\vec{Q}\cdot\vec{r}) \qquad (20)$$

provided that the wave-function of the mobile electrons within the interlayer is given by the Eq. (17) we can easily determine the spectrum of electrons as

$$\begin{aligned}
&\varepsilon_k(Q) \approx \left\langle \Phi_{CDW}(\vec{Q},\vec{r})\middle|H(r)\middle|\Phi_{CDW}(\vec{Q},\vec{r})\right\rangle = \\
&= -\frac{\hbar^2}{2m}\left\langle \Phi_{CDW}(\vec{Q},\vec{r})\middle|\nabla^2\middle|\Phi_{CDW}(\vec{Q},\vec{r})\right\rangle + \\
&+\left\langle \Phi_{CDW}(\vec{Q},\vec{r})\middle|V_0\middle|\Phi_{CDW}(\vec{Q},\vec{r})\right\rangle + \qquad (21)\\
&+\left\langle \Phi_{CDW}(\vec{Q},\vec{r})\middle|\chi os(\vec{Q}\cdot\vec{r})\middle|\Phi_{CDW}(\vec{Q},\vec{r})\right\rangle = \ldots \\
&\ldots = \frac{\hbar^2}{2m}\left(\vec{k}^2 + 2\left(\vec{k}^2 + \vec{Q}^2\right)|\alpha|^2\right) + V_0\left(1 + 2|\alpha|^2\right) + G\alpha
\end{aligned}$$

where $k^2 = k_{\|}^2 + k_{\perp}^2$, and it is assume that $\alpha_+ = \alpha_- = \alpha$.

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