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## ON BIQUADRATIC INTERLAYER COUPLING IN MAGNETIC MULTILAYERS

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Intrinsic biquadratic interlayer coupling in magnetic layered structures is analysed theoretically within the spin-polarized free-electron approximation. The corresponding coupling parameter oscillates with the interlayer thickness with the oscillation period equal to half of that for bilinear term. An extrinsic mechanism following from a competition between the intra- and interlayer exchange coupling is also discussed.

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Experimental data on interlayer exchange coupling in artificially layered magnetic structures [1-3] can be accounted for if one assumes the coupling energy per unit area in the following phenomenological form:

$$E_i = -J_1 \frac{\mathbf{M}_1 \cdot \mathbf{M}_2}{M_1 M_2} - J_2 \left( \frac{\mathbf{M}_1 \cdot \mathbf{M}_2}{M_1 M_2} \right)^2 = -J_1 \cos \theta - J_2 \cos^2 \theta, \quad (1)$$

where  $M_1$  and  $M_2$  are the magnetizations of both films at the interlayer,  $\theta$  is an angle between the magnetizations, and  $J_1$  and  $J_2$  are respectively the bilinear and biquadratic coupling parameters. The bilinear term behaves oscillatory-like with increasing interlayer thickness and is essentially well understood. The origin of biquadratic term is not so clear. Slonczewski [4] proposed a mechanism of the biquadratic term, which follows from a competition between the ferromagnetic intra-layer exchange coupling and interlayer bilinear coupling in the presence of interface roughness (steps). This mechanism, however, does not contribute in the case of ideal interfaces.

Here we show that the biquadratic coupling is an intrinsic feature of metallic magnetic layered structures and exist in the absence of interface roughness [5, 6]. The interlayer coupling cannot be written in a simple Heisenberg-like form, but includes also higher order terms. This follows from the fact that the electronic wave functions depend on the relative orientation of the film magnetizations. Consequently, the coupling parameter also depends on the angle between the magnetizations, which essentially is equivalent to the occurrence of higher order terms. Such intrinsic mechanism can be obtained within any model of magnetic systems provided appropriate electronic states are taken into account.

Consider, for example two ferromagnetic films of thickness  $D$  separated by a non-magnetic spacer of thickness  $d$ . For simplicity, we restrict considerations to spin-polarized free-electron approximation. If the chemical potential  $\mu$  is constant, then the parameters of bilinear and biquadratic terms can be calculated from the formulae

$$J_1 = \frac{1}{2} (\Omega_{\uparrow\downarrow} - \Omega_{\uparrow\uparrow}), \quad (2a)$$

$$J_2 = \Omega_{\perp} - \frac{1}{2} (\Omega_{\uparrow\downarrow} + \Omega_{\uparrow\uparrow}), \quad (2b)$$

where  $\Omega_{\uparrow\downarrow}$ ,  $\Omega_{\uparrow\uparrow}$  and  $\Omega_{\perp}$  are the thermodynamic potentials per unit area for the antiparallel ( $\theta = \pi$ ), parallel ( $\theta = 0$ ) and perpendicular ( $\theta = \pi/2$ ) orientations of the film magnetizations. If however the total number of  $s$  electrons is conserved then the parameters  $J_1$  and  $J_2$  can be found from

$$J_1 = \frac{1}{2} (E_{\uparrow\downarrow} - E_{\uparrow\uparrow}), \quad (3a)$$

$$J_2 = E_{\perp} - \frac{1}{2} (E_{\uparrow\downarrow} + E_{\uparrow\uparrow}), \quad (3b)$$

where  $E_{\uparrow\downarrow}$ ,  $E_{\uparrow\uparrow}$  and  $E_{\perp}$  denote the total energy of the electron system in the corresponding configuration.

The thermodynamic potentials  $\Omega_{\uparrow\downarrow}$ ,  $\Omega_{\uparrow\uparrow}$  and  $\Omega_{\perp}$  (or the energies  $E_{\uparrow\downarrow}$ ,  $E_{\uparrow\uparrow}$  and  $E_{\perp}$ ) are determined by the corresponding spectra of single-electron states which can be found from the Schrödinger equation

$$\left[ -\frac{\hbar^2}{2m} \nabla^2 + U(x) + \mu_B h(x) \sigma \right] \Psi(r) = E \Psi(r), \quad (3)$$

where  $U(x)$  (axis  $x$  is normal to the structure) is the potential energy,  $h(x)$  is a molecular field in the ferromagnetic material,  $\sigma$  is the Pauli spin operator and  $\Psi(r)$  is the electron wave function in the spinor form. We assume that  $U(x) = U_0$  and  $|h(x)| = h_0$  inside both ferromagnetic films, whereas  $U(x) = 0$  in the interlayer (where also  $|h(x)| = 0$ ).

Results of appropriate numerical calculations are shown in Fig. 1 and Fig. 2, where for comparison both  $J_1$  and  $J_2$  are shown as a function of the interlayer thickness  $d$ . For simplicity we assumed there  $U_0 = \mu_B h_0$ . Figure 1 shows the curves for a constant chemical potential  $\mu$ , whereas Fig. 2 — for a constant number of  $s$  electrons. The parameter of biquadratic coupling oscillates with a period  $\Lambda/2$ , where  $\Lambda$  is the oscillation period for the bilinear term. Its magnitude — for parameters assumed in numerical calculations — is smaller approximately by a factor of 5 in comparison with the corresponding bilinear parameter. In Fig. 2 the coupling parameter is slightly larger in comparison with that in Fig. 1 and there is also some phase shift between those two curves.

In the model considered here the parameter  $J_2$  vanishes at points where also  $J_1 = 0$ . This, of course, cannot account for the experimentally observed perpendicular orientation of the film magnetizations. However, one has to realize that in experimental situations there is always a negative Slonczewski-type contribution to  $J_2$ . This contribution, when added to the one calculated above, leads to regions where  $J_2$  dominates and  $J_2 < 0$ . There is also another negative contribution to

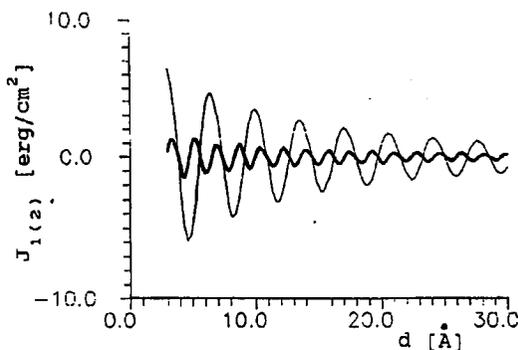


Fig. 1. Bilinear  $J_1$  (thin line) and biquadratic  $J_2$  (bold line) coupling parameters as a function of the interlayer thickness calculated for  $U_0 = \mu_B h_0 = 0.75$  eV,  $U_1 = 0$ ,  $D = 5$  Å and for the constant chemical potential  $\mu = 3$  eV.

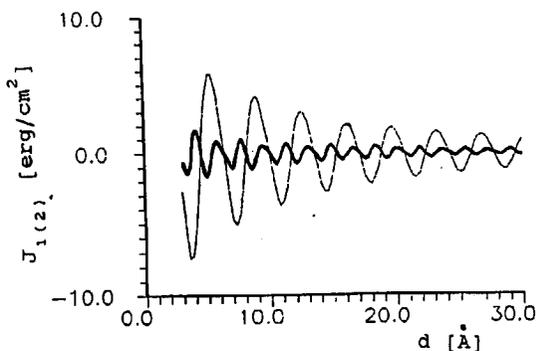


Fig. 2. The same as in Fig. 1 but for a constant particle number determined by the chemical potential in the bulk limit  $\mu = 3$  eV. The other parameters are the same as in Fig. 1.

$J_2$  which exists also for ideally flat interfaces (when Slonczewski's contribution vanishes). It results from a competition between the bilinear interlayer coupling and the intra-layer exchange interaction when the former penetrates sufficiently deeply into the magnetic films [6]. Its origin can be explained easily within a simple RKKY-type model of the coupling between localized magnetic moments. Let us assume that each magnetic monolayer adjacent to the interlayer interacts across the interlayer with two nearest atomic planes of the second magnetic film, with  $I_1$  and  $I_2$  being the relevant coupling parameters. For simplicity we will neglect coupling between further magnetic atomic planes. Let the direct intra-layer exchange coupling between two atomic planes at the interface be described by the parameter  $I$ . If  $I_1$  and  $I_2$  are of opposite signs then the two interface atomic planes of each magnetic film are subject to opposite torques which drive them to align oppo-

sitely. However, the intra-layer exchange coupling, which is usually much stronger than the interlayer one, resists the torques keeping them almost parallel. Taking into account classical expressions for inter- and intra-layer exchange coupling one may show that the total energy can be written in the form (1) with the parameters  $J_1$  and  $J_2$  given by the formulae

$$J_1 = I_1 + 2I_2 \quad (5a)$$

and

$$J_2 = -I_2^2/I. \quad (5b)$$

In the limit of rigid film magnetization ( $I \rightarrow \infty$ ) the total energy of the interlayer coupling reduces to the linear term only. If, however,  $I$  is finite, then an additional biquadratic term appears as a result of the competition between the inter- and intra-layer exchange interactions. In a more accurate description one should consider a coupling between further atomic planes and use more realistic models or calculate the coupling parameters from the first principles [7]. However, the presented model — despite of its simplicity — accounts qualitatively for all basic features of the biquadratic term in the interlayer coupling.

The considerations presented here apply to ultra-thin films, where the quantum size effects are important. This takes place when the electron mean free path is of the order of or larger than the structure thickness. Most of the experiments correspond rather to the opposite limit, where the mean free path is small in comparison to the sample thickness. In that case the results presented above are not applicable and one may also expect different qualitative behaviour. The influence of a metallic substrate was neglected in the above calculations. However, it can also play a significant role.

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