**The enhancement of low-temperature excitation of magnons via interlayer exchange coupling in perpendicularly magnetized [Co/Pd] multilayers**

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**Abstract**

In this study we analyze the correlation between magnetization and magnetoresistance of perpendicularly anisotropic [Co/Pd] multilayered films with different thickness of Pd layers *t*Pd = 0.6-2.0 nm in a wide range of temperatures *T* = 4-300 K. We revealed that electron scattering on magnons makes a significant contribution to the magnetoresistance of the multilayers regardless of the layer thickness. Contrary to expectations, the effect of magnon magnetoresistance (MMR) increases with decreasing temperature below *T* = 50 K in the films with *t*Pd = 0.8 and 1.0 nm. The revealed low-temperature MMR increase, which is most pronounced in the [Co0.5/Pd1.0] multilayers, is associated with the enhanced magnon excitation due to antiferromagnetic exchange coupling between the Co layers. The latter ensures an atypical shape of the magnetization curves of the [Co0.5/Pd1.0] multilayers at low temperature in perpendicular magnetic field, which combine a quadratic hysteresis loop of a perpendicularly anisotropic ferromagnet and an anomalous magnetization drop resulting from a violation of the ordering of magnetic moments and their amplified oscillations initiated by the interlayer exchange coupling.

The mechanisms of resistivity in ferromagnetic (FM) metals are significantly more diverse as compared to normal metals (NM) due to electron scattering on magnetic moments. In addition to the spin-dependent scattering of conduction electrons on localized magnetic moments of ferromagnet, providing anisotropic, domain wall, giant, etc. magnetoresistance (MR) mechanisms, their spin-flip scattering on the oscillations of magnetic moments ensures an additional magnon mechanism contributing to MR [1]. Magnons, quasiparticles of spin waves, associated with the collective precession of spin moments in magnetically ordered systems have recently attracted widespread attention due to their potential application for the low-energy, high-speed and high-frequency information transfer and processing [2, 3]. The approaches to the efficient excitation, manipulation, propagation, control and detection of magnons in low-dimensional materials and nano-devices are core issues in spintronics and magnonics [3, 4]. The magnon magnetoresistance (MMR), which consists in high-field damping of thermally-excited spin waves in *3d* FM metals [5, 6], is a sensitive parameter that makes it possible to detect magnons and estimate their contribution to the electron scattering both in bulk materials and thin ferromagnetic films and nanostructures [5-9]. In this Letter we discuss the enhanced low-temperature excitation of magnons provided by the antiferromagnetic (AFM) interlayer exchange coupling (IEC) in [Co/Pd] multilayers (MLs), which is observed through an increase in the magnon contribution to their MR. We have previously reported the detected propagation of spin waves and pronounced MMR in the [Co/Pd] MLs [10-12]. Despite the fact that spin waves are generated within the Co FM layers, the crucial role of Pd was demonstrated, since the contribution of MMR is significantly reduced and is practically undetectable in similar [Co/Pt] MLs, in which the Pd layers are replaced by Pt layers of the same thickness [13]. Here, we demonstrate the sensitivity of spin waves to the magnetic interaction between the FM layers within multilayered structure, which allows detecting their AFM coupling even through ultrathin NM layer that is masked by other type interactions and unavailable for revealing by standard techniques, but can be distinguished by evaluating the MMR contribution to magnetoresistance.

The Ta5 nm/Pd3 nm/[Co0.5 nm/Pd*t*nm]х5/Co0.5/Pd3 nm/Ta5 nm thin films analyzed in this study were deposited on thermally oxidized flat Siwafers using ultra-high vacuum magnetron sputtering system (AJA International, Inc., USA). The thickness of Pd layers (*t*Pd) was varied in the range of 0.6-2.0 nm. The top Pd/Ta bilayer was used to prevent oxidation of the MLs, while the bottom Ta/Pd bilayer serves to promote strong (111) texture and pronounced perpendicular magnetic anisotropy (PMA) of the [Co/Pd] MLs. The layer thicknesses were determined from the deposition time and calibrated deposition rates and verified using high-resolution transmission electron microscopy for several compositions [13]. The magnetic properties were characterized using the vibrating sample magnetometry (VSM) option of the Quantum Design Physical Property Measurement System (PPMS) in the temperature range *T*= 4-300 K under an applied magnetic field *H* up to 90 kOe. The MR of the films was measured by a standard four-probe method using the direct current (*I* = 5 mA) resistivity option of PPMS. The field dependences of MR(*H*) were obtained in magnetic field *H* applied along the film normal.

Magnetization curves *M*(*H*) of the [Co0.5/Pd1.0] and [Co0.5/Pd2.0] MLs with strong PMA are similar at room temperature (RT) in the field applied along their easy axis, but differ significantly at low temperatures below *T* = 100 K, as illustrated in Fig. 1(a)-(b). Particularly, the magnetizationof the [Co0.5/Pd1.0] MLs decreases with decreasing field, starting from the anisotropy field *H*A ~ 35 kOe (lower inset to Fig. 1(a)) down to approximately zero field, and regains its value corresponding to the saturation magnetization *M*S only when the magnetic field alters its direction to the opposite (upper inset to Fig. 1(a)). That is, an anomalous drop in the *M*S value is characteristic of this MLs in the field range of ~0-35 kOe at *T* = 4-50 K. It should be mentioned that such a peculiarity of magnetization has not been described previously for magnetic films or MLs. The magnetization curves of the [Co0.5/Pd2.0] MLs, oppositely, demonstrate an extra increase in their low-temperature *M*S value provided by an additional superparamagnetic (SP)-like contribution to *M*(*H*) (Fig. 2(b)). This increase exceeds the commonly observed growth of *M*S due to the damping of the temperature-induced fluctuations of magnetic moments according to the Bloch’s law (inset in Fig. 2(b)) and is associated with the progressively increasing Pd polarization at low temperatures [14-16].



Figure 1. (a)-(b) Magnetization curves *M*(*H*) of [Co0.5/Pd1.0] MLs (a) and [Co0.5/Pd2.0] MLs measured at different temperatures *T* = 4-300 K in a perpendicular magnetic field *H*; upper insets to (a)-(b) show enlarged central parts of the *M*(*H*) curves; lower inset to (a) shows the *M*(*H*) curves of [Co0.5/Pd1.0] MLs measured at *T* = 4 K in a magnetic field applied both along the film normal and in the film plane; lower inset to (b) demonstrates temperature dependences of saturation magnetization *M*S(*T*) of [Co0.5/Pd1.0] and [Co0.5/Pd2.0] MLs. (c) Temperature dependences of coercive fields *H*C(*T*) of [Co0.5/Pd*t*] MLs with different Pd thickness *t*Pd = 0.6-2.0 nm; inset to (c) shows the thickness dependence *H*C(*t*Pd) of [Co0.5/Pd*t*] MLs for *T* = 4 and 300 K.

The observed difference in magnetization of the above films with different *t*Pd thickness is accompanied by a non-monotonic dependence of the coercive field *H*C on *t*Pd (Fig. 1(c)). Fig. 1(c) illustrates *H*C(*T*) dependences of the films with different *t*Pd normalized to their *H*C values at RT, while inset shows the absolute *H*C values at RT and at *T* = 4 K as a function of *t*Pd. The oscillating character of the *H*C(*t*Pd) dependence is clear from the figure, with the amplitude of oscillations increasing at low temperature. The oscillations in *H*C(*t*Pd) are supposed to arise due to the oscillations in the interaction between the Co layers with changing thickness of the Pd layers between them. Indeed, such oscillations in strength and type (FM or AFM) of coupling are characteristic of the FM/NM/FM trilayers (RKKY coupling [17]). However, the joint switching of magnetization of the FM layers in the studied MLs does not allow us to reliably identify the type of their interaction from the magnetization curves, since IEC is often masked by FM interactions of other types [16, 17]. The indirect confirmations of oscillating RKKY interaction in the FM/NM/FM trilayers and FM/NM superlattices can be changes in their *H*C value [14] or the giant MR effect [18] on the NM spacer thickness. However, distinguishing the mechanisms of interlayer coupling is more complicated in the systems with an easily polarizable NM, such as Pd or Pt. Since the RKKY interaction originates in spin polarization of conduction electrons, the FM-proximity induced polarization in Pd or Pt influences the interlayer coupling, shifting it towards FM interaction [14]. Therefore, the manifestation of the AFM-IEC interaction in FM/Pd(Pt)/FM systems is debatable [14, 17], but the oscillatory behavior of the strength of exchange coupling between the FM layers with a variation of NM thickness was clearly identified using ferromagnetic resonance [16, 19]. It is noteworthy that the first local minimum of the strength of interlayer exchange observed for the Fe/Pd(001)/Fe trilayer corresponds to 6 atomic monolayers of Pd (~1.15 nm).

In line with this, the [Co0.5/Pd1.0] MLs, which show a low-temperature drop of magnetization (Fig. 1(a)), has minimum *H*C value (Fig. 1(c)). Simultaneously, a pronounced low-temperature increase in the *H*C(*T*) value of the [Co0.5/Pd2.0] MLs (Fig. 1(c)) correlates with the corresponding increase in its *M*S value (lower inset to Fig. 1(b)) and indicates the intensifying the FM-IEC between Co layers, related to the Pd polarization in this MLs. On the contrary, the Pd polarization in the [Co0.5/Pd1.0] MLs is clearly less pronounced (lower inset to Fig. 1(b)). Therefore, the observed low-temperature peculiarities of magnetization of [Co0.5/Pd1.0] MLs can potentially be attributed to the manifestation of AFM-IEC, which competes with FM interaction realized via direct FM exchange (through pinholes, enhanced by Pd polarization) or magnetostatic mechanisms [17, 20] and enforces antiparallel alignment of the magnetization of the FM layers. However, the AFM interaction is apparently not strong enough to provide complete AFM ordering of MLs, bur may be capable of causing some specific configurations of magnetic moments, for example, their partial antiparallel alignment or tilted orientation that ensures the described drop in magnetization (Fig. 1(a)). It should be mentioned that the minimum magnetization value (*M*min), which characterizes this drop, does not fall below the RT value of *M*S, i.e. the magnitude of the described effect of low-temperature magnetization drop does not go beyond the decrease in magnetization due to its temperature-induced fluctuations at RT.

As was established earlier, [Co/Pd] MLs are characterized by pronounced propagation of spin waves, which has a strong effect on their magnetoresistance [10-12]. The MR(*H*) curves of [Co/Pd] MLs with different compositions, namely [Co0.5/Pdt] MLs with *t*Pd = 0.8, 1.0 and 2.0 nm and [Co*t*/Pd1.0] MLs with *t*Co = 0.4 and 0.5 nm are shown in Fig. 2(a)-(d). The MR curves in Fig. 2 are obtained after extracting the ~*H*2 contribution of Lorentz MR.



Figure 2. (a)-(d) Field dependences of magnetoresistance MR(*H*) of [Co0.5/Pd*t*] MLs with *t*Pd = 2.0 nm (a), 1.0 nm (b), and 0.8 nm (c) and [Co*t*/Pd1.0] MLs with *t*Co = 0.5 nm (b) and 0.4 nm (d) measured at different temperatures *T* = 4-300 K; color lines with scatters show experimental MR(*H*) curves after extracting the Lorentz contribution to MR, black solid lines illustrate the approximation of MR(*H*) curves according to the MMR mechanism, and black arrows indicate changes in the MR effect with decreasing temperature; insets to (a)-(d) show enlarged central parts of the experimentalMR(*H*) curves. (e) Temperature dependences of pre-factor *T*/*D*2 in the approximation of MR(*H*) curves within the framework of the MMR mechanism associated with to the ratio of MMR effect for the above films. (d) Temperature dependences of experimental (red scatters) and calculated for Co (black scatters) exchange stiffness constant *D* normalized to zero-temperature stiffness *D*0 of Co (left axis) and temperature dependence of saturation magnetization *M*S(*T*) (right axis) of [Co0.5/Pd2.0] MLs.

The shape of the MR(*H*) curves of all studied MLs perfectly corresponds to the MMR mechanism, which is confirmed by a good agreement with the fitting curves according to the following relation (black lines in Fig. 2(a)-(d)):

, (1)

where Δρ corresponds to the change in resistivity due to MMR, *D*(*T*) = *D*0(1-*d*1*T2*) is the exchange stiffness constant, *D*0 is zero-temperature stiffness of spin-waves, μB is the Bohr magneton, and *k* is the Boltzmann constant [5, 7].

The main peculiarity of the magnon MR mechanism revealed in the studied films is a weak damping of spin waves with decreasing temperature, which are typically negligible below *T* = 77 K for both bulk and thin film Co [5-7]. The latter is evidenced in non-vanishing pre-factor *T*/D2 in (1) (Fig. 2(e)), which is proportional to the contribution of electron scattering on magnons to MR. The temperature dependence of the exchange stiffness constant *D*(*T*), estimated from the *T*/*D*2 fitting parameter, differs significantly from that for the calculated *D* value for Co (*D*0 = 435 meV Å2; *d*1 = 1.5×10-6 K-2 [5]), as illustrated in Fig. 2(f) for the [Co0.5/Pd2.0] MLs. Such a difference is supposed to be due to the excitation of fluctuations of polarized magnetic moments in the Pd layers [16]. In doing so, the low-temperature polarization of Pd and its mediating the interlayer coupling between Co layers provides the formation of quasi-uniform magnetic phase with the reduced stiffness of the excited oscillations (spin-waves), as compared to the pure Co. The latter is confirmed by the correlation of the temperature dependences of *D* and *M*S (Fig. 2(f)), with the stiffness continuously decreasing with decreasing temperature in accordance with the increasing fraction of polarized Pd atoms. Simultaneously, the magnetic phase formed in the [Co0.5/Pd2.0] MLs are characterized by significantly reduced Curie temperature *T*C ~ 770 K, estimated from the approximation of its *M*S(*T*) dependence according to the Bloch’s *T*3/2 law (lower inset to Fig. 1(b)), as compared to that of the [Co0.5/Pd1.0] MLs (~1260 K), which is much closer to that of Co (~1388 K) [11]. The latter confirms the “dilution” of the magnetic phase formed in the [Co/Pd] MLs by the magnetic moments of Pd atoms.

In addition to the weakly damped spin-waves in the studied MLs, the films with *t*Pd = 0.8 and 1.0 nm show unexpected increase in the MMR effect at *T* < 50 K (arrows in Fig. 2(b)-(c)), with an increase being maximum for the [Co0.5/Pd1.0] MLs. Its MMR effect at *T* = 4 K becomes equal to that at RT. Such a peculiarity is evident both from the slope of the low-field MMR curves (insets to Fig. 2(a)-(d)) and from the increased *T*/*D*2 parameter (Fig. 2(c)). It is noteworthy that the temperature range of the MMR effect growth corresponds to that of manifestation of the low-temperature drop in magnetization of the [Co0.5/Pd1.0] MLs (Fig. 1(a)). Moreover, the temperature dependences of both revealed peculiarities of the *M*(*H*) and MR(*H*) curves are found to correlate with each other. The latter is illustrated in Fig. 3. The drop of magnetization due to the supposed AFM-IEC is characterized here by the *M*min value, while the low-temperature increase in spin-wave excitation is defined by the *T*/*D*2 parameter associated with the MMR contribution. The revealed correlation seems to be convincing confirmation that both effects have the same origin.



Figure 3. Temperature dependences of a decreased magnetization *M*min due to IEC (a) and the MMR effect ratio *T*/*D*2 (b) of the [Co0.5/Pd1.0] MLs; inset demonstrates the definition of the *M*min and *M*S parameters from magnetization curve.

The increase in the magnon contribution to MR with decreasing temperature is a nontrivial effect, since besides artificial generation of spin waves (spin pumping), temperature-induced fluctuations of magnetic moments are considered to be a main source of magnons. On the other hand, the sensitivity of spin fluctuations to the RKKY coupling has been previously reported both for the spin waves generated in the interacting FM layers (interference between the pumped spin currents from two magnetic layers [4]) and within the polarized Pd spacer [16]. Taking this into account, we can assume that the AFM-IEC provides the low-temperature increase in magnons excitation in the [Co0.5/P1.0] MLs, which in turn decreases the magnetization and contributes to the increased MMR effect. The scalability of the detected effects of increasing MMR (Fig. 2(e)) and decreasing magnetization (Fig. 1(a)-(b)) for the MLs of different layer thicknesses indicates the high sensitivity of magnons to interlayer coupling.

In conclusion, we have revealed the enhanced MMR mechanism of electron scattering and deviation of the shape of magnetization curves from typical quadratic hysteresis loops for the [Co/Pd] MLs with several thicknesses of Pd layer at low temperatures *T* ≤ 50 K. It is shown that the found peculiarities of the *M*(*H*) and MR(*H*) dependences oscillate with the Pd thickness that discloses their origin, which lies in the interlayer exchange coupling of Co layers. The convincing correlation between the identified low-temperature enhancement of the MMR effect and drop of magnetization indicates sensitivity of spin waves generated in the [Co/Pd] MLs to AFM-IEC. This opens up new opportunities for magnons generation and manipulation even in films and multilayers with extremely thin FM layers.

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**Author Declaration**

***Conflict of Interest Statement***

The authors have no conflicts to disclose.

***Author Contributions***

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**Janusz Przewoźnik:** Data curation, Investigation, Resources,Writing/Review & Editing.

**Czesław Kapusta:** Data curation, Investigation, Resources,Writing/Review & Editing.

**Ivan Svito:** Data curation, Investigation, Resources.

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**Johan Åkerman:** Writing/Review & Editing.

**Thi Ngoc Anh Nguyen:** Conceptualization, Methodology, Writing/Original Draft Preparation.

**Data Availability Statement**

The data that supports the findings of this study are available from the corresponding author upon reasonable request.

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