Oscillating behavior of magnetization in Pd/Fe and dilute $Pd_{1-x}Fe_x$/Fe multilayers

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$Pd_{1-x}Fe_x$/Fe multilayers with $x=0$, 0.034, 0.056, 0.071 have been prepared by magnetron sputtering. The measurement of saturation magnetization in Pd/Fe ML's in the range of Pd thickness from 4 to 18 ML shows that the interlayer couplings are ferromagnetic and M_s possesses a long-range oscillation with a period of 4 ML Pd. The oscillation phase and period in $Pd_{0.966}Fe_{0.034}$/Fe and $Pd_{0.944}Fe_{0.056}$/Fe multilayers remain as compared with their reference Pd/Fe multilayers. A discussion is given about the discrepancy between theory and experiments, concerning whether a negative polarization of Pd in the inner spacer and a long-range ferromagnetic oscillation appear in Pd/Fe systems. [S0163-1829(99)01418-6]

I. INTRODUCTION

During the last decade, the oscillatory coupling between two ferromagnetic (F) layers separated by a nonferromagnetic metallic spacer has been one of the most discussed phenomena,[1–5] since Grünberg *et al.* found the antiferromagnetic (AF) interlayer coupling in the Fe/Cr/Fe sandwich.[1] It is confirmed that antiferromagnetic coupling is closely related to the Fermi-surface character of the nonferromagnetic spacer. On the other hand, attention has been paid to the layered structure with the near ferromagnetic material spacer Pd, Pt, and an appreciable induced polarization in the Pd layer was found in Pd/Fe.[6–12]

Celinski *et al.* synthesized Fe(001)/Pd_n(001)/Fe(001) sandwiches and found experimentally some interesting results (a) when $n=4$, Pd is ferromagnetic polarized by Fe and a magnetic moment of $0.25\mu_B$ per Pd atom in the ferromagnetically ordered Pd is deduced,[6] which agrees well with the theoretical calculation given by Blügel *et al.*;[11] (b) for $n=12$, the ferromagnetic interlayer coupling shows an oscillation with a period of 4 ML, and there exists a crossover from F to AF coupling at 12 ML Pd.[9] Childress *et al.*[7] investigated then (001)Fe/Pd superlattices with the same grown mode as that of Celinski and they found no evidence of AF coupling between Fe layers in the range of $5\leqslant n\leqslant 25$ and a constant average value of M_s of the (001)Fe/Pd superlattice at 15 K in the whole Pd thickness.

More recently, Stoeffer *et al.* have theoretically investigated the magnetic properties of Fe/Pd superlattice.[13] They considered two model structures for the Pd spacer: (1) a fct structure for which the Pd atoms keep their bulk atomic volume (CAV) and (2) a fcc structure for which the Pd atomic volume is expanded (EAV). The calculated mean magnetic moment per Pd atom for Fe_3Pd_4 in the EAV structure equals $0.265\mu_B$, which is in agreement with the experimental result given by Celinski *et al.*, but it is much smaller than the result of Childress *et al.*[7] The Pd polarization for the CAV structure is limited to the interfacial atomic layer, while the Pd spacer is entirely polarized up to $n=15$ for the EAV one. It is not consistent with the result of Celinski *et al.* that the whole Pd layer is in ferromagnetic order just for $n_{Pd}=4$.[9]

Li *et al.* investigated the magnetic properties of the sputtered (20 Å)Fe/(t Å)Pd ML's in the range of $t=6-60$ Å. They found that the saturation magnetization M_s of Fe at 5 K is enhanced and shows an oscillationlike behavior with the increase of t_{Pd}, i.e., a negative polarization of Pd atoms in the inner Pd layer appears.

So far we can see that there are some discrepancies between the theory and experiments and between the differential experimental results as outlined above. The main points are concerned with whether a negative polarization of Pd atoms in the inner Pd spacer layers appears when n_{Pd} is larger than 4 and whether a long-range ferromagnetic oscillation exists in Pd/Fe systems.

In order to attempt to resolve these discrepancies we measured M_s as a function of Pd thickness for the sputtered Pd/Fe multilayers. Moreover, we also measured M_s as the function of the PdFe layer thickness in the sputtered $Pd_{1-x}Fe_x$/Fe ($x=0.034,0.056,0.071$) multilayers in order to understand the effect of the existence of Fe impurity on the magnetic properties of Pd/Fe multilayers.

II. EXPERIMENTS

The multilayers of a 30 nm thick $Pd_{1-x}Fe_x$ buffer layer/ t Å $Pd_{1-x}Fe_x$/30.1 Å Fe in the range of $t=11-41.6$ Å for $x=0$,0.034,0.056,0.071, respectively, were prepared on a 0.2 mm thick glass substrate by magnetron sputtering with Ar gas at 7.5 mTorr. The base vacuum was about 4×10^{-7} Torr. The substrate holder was cooled by water during sputtering. The thickness of each layer was controlled by the sputtering time through a preset computer program after fixing the other sputtering parameters. The number of the period is 20 for all the prepared samples. The addition of Fe into the Pd layer was carried out by means of increasing the area ratio of Fe to Pd on a composite target. The Fe concentration in the Pd spacer layer was determined by an inductively coupled plasma-atomic emission spectrometry. The modulation character and crystal structure of the samples were analyzed by small- and large-angle x-ray diffraction (XRD). The saturation magnetization M_s was measured by means of an altering-gradient magnetometer with the applied magnetic field in the film plane at room temperature. Con-

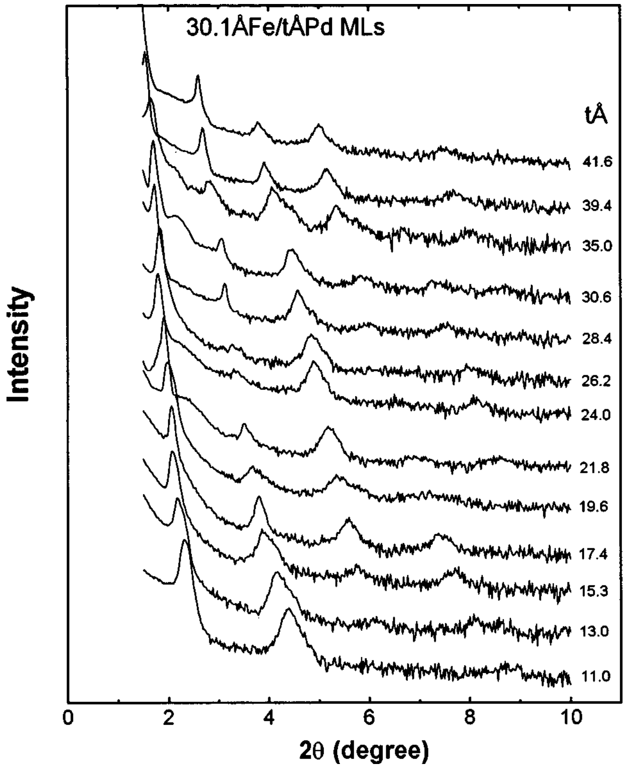

FIG. 1. Small XRD patterns for the 30.1 Å Fe/t Å Pd multilayers in the range of $t_{Pd}=11.0$ to 41.6 Å.

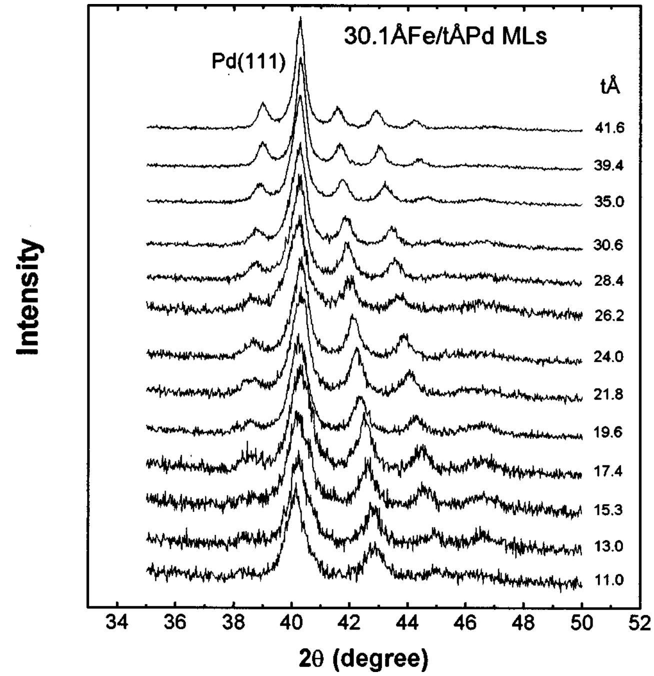

FIG. 2. Large XRD patterns for the 30.1 Å Fe/t Å Pd multilayers in the range of $t_{Pd}=11.0$ to 41.6 Å.

sidering the error of determining the Fe layer thickness and the surface area, the general error of M_s is less than about 5%. According to the measured hysteresis loops we can confirm that the magnetic moments of the adjacent Fe layers are parallel in all samples.

III. RESULTS AND DISCUSSION

Figures 1 and 2 show the small- and large-angle XRD patterns for the t Å Pd/30.1 Å Fe ML's in the range of $t=11.0-41.6$ Å, respectively. Two or four small-angle XRD peaks can be clearly seen from the measurements. It specifies that the prepared samples possess a good layered structure. For the large-angle XRD measurements, the Pd (111) peak at $2\theta=40.1°$ is pronounced for all the samples and there are also three or more superlattice peaks on each curve. The Pd (111) peak which occurred at $2\theta=40.1°$ coincides with its bulk value, therefore we speculate that the sputtered Pd/Fe ML's do not like the superlattice prepared by molecular-beam epitaxy (MBE), where a lattice expanded Pd(001) in ultrathin Fe(001) appears.[6–9] After Fe doping, the layered structure that the Pd/Fe ML's possess remains and the position of all XRD peaks has not been changed. Moreover, no Fe or Fe alloy peaks were found in the large XRD measurements.

Figure 3 shows the average saturation magnetization M_s on the total Fe layers in the sample as the function of the thickness of Pd layer t_{Pd}. First, we can see that the interlayer exchange couplings are ferromagnetic, on the whole Pd thickness range that can be also confirmed from the hysteresis loop measurements. It is not in agreement with the result given by Celinski *et al.* but in agreement with the results of Childress *et al.* and Bloemen *et al.*[8] Then, we can see from this curve that an obvious oscillation of M_s with the increase of t_{Pd} appears and the oscillation period is about 4 ML. This value is the same with the result of Celinski *et al.*[9] measured from the interlayer exchange coupling constant J as the function of the thickness of Pd layer in 5.7 ML Fe/t ML Pd/9.6 ML Fe trilayers ($t=5-18$) that is from Fig. 3 for the comparison. In the inset of Fig. 3, we also quote the data about M_s as a function of t_{Pd} given by Li *et al.*[15] There

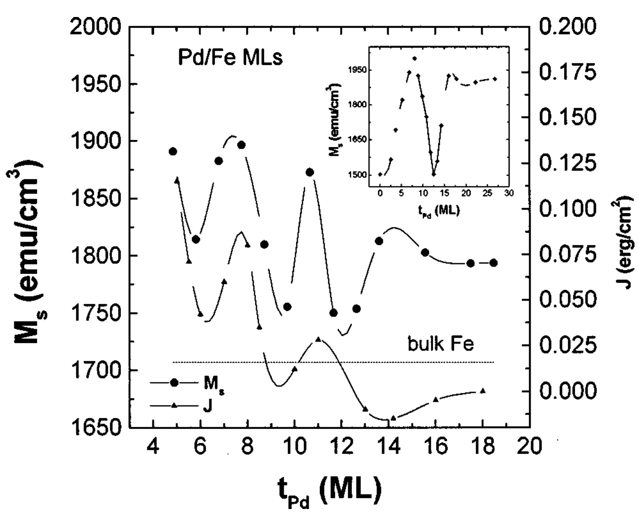

FIG. 3. Saturation magnetization M_s averaged on the Fe layers as the function of the Pd thickness t_{Pd}, for the 12 ML Fe/t ML Pd multilayers. The curve of J vs t_{Pd} is from Ref. 7 and the data shown in the inset is from Ref. 15.

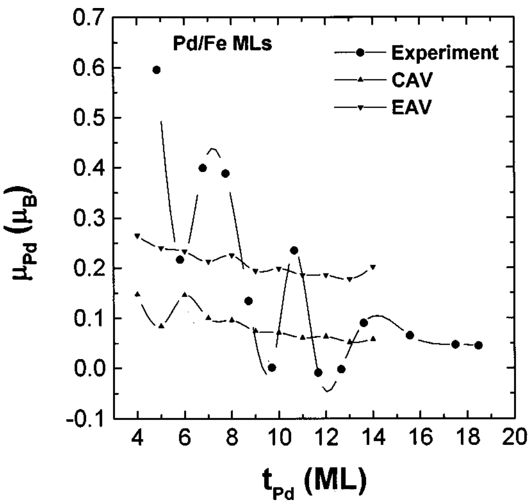

FIG. 4. Averaged value of μ_{Pd} as the function of the Pd thickness t_{Pd} for the 12 ML Fe/t ML Pd multilayers. The theoretical results of μ_{Pd} as the function of t_{Pd} for CAV and EAV are from Ref. 13.

is only a broad peak of M_s below about 12.5 ML of the Pd thickness. Obviously, this result is different as compared with ours.

As mentioned above, M_s is larger than the value of Fe bulk. The enhancement of the magnetization is expected from the spin polarization of Pd layers. For a further elaborating, we subtract the contributions of Fe layers ($\mu_{Fe} \approx 2.2\mu_B$) and the buffer layer from the total measured magnetization M_s and average the remains on the total Pd atoms. Figure 4 gives this average value μ_{Pd}. One can find that μ_{Pd} is the same as M_s and shows an oscillation with the increase of the Pd thickness, so that we can attribute the oscillation of M_s to the Pd polarization in the Pd/Fe ML's. The oscillation of M_s or μ_{Pd} indicates the existence of a negative polarization of Pd in the inner spacer and this negative polarization is different from the theoretical results for Pd having CAV and EAV structures[13,14] that are quoted in Fig. 4, respectively. From the quoted curves, we can see that the Pd polarization exists just in the interface for the CAV case and its polarization remains unchanged for the EAV one.

As for the comparison, our results are much more similar to the results of Celinski *et al.* because we both obtained the interlayer oscillatory coupling with the same period and phase and found the existence of the negative polarization of Pd in the inner spacer layer, except for the crossover from F to AF coupling at $n=12$ give by Celinski *et al.* As for the Pd polarization, Celinski *et al.* have done the ferromagnetic resonance measurement in 5 ML Fe/4 ML Pd/10 ML Fe trilayers and showed that the Pd/Fe interface contributed $0.9\mu_B$. Assume that the interface Fe has the increase in magnetic moment $0.4\mu_B$, then the remaining $0.5\mu_B$ belongs to two Pd layers in the Pd/Fe interface. Distributing this moment evenly between Pd layers results in a magnetic moment of $0.25\mu_B$/Pd atom. In the case of our sputtered 12 ML Fe/5 ML Pd ML's, $\mu_{Pd} \approx 0.6\mu_B$ is deduced and it is obviously larger than the $0.25\mu_B$/Pd atom given by Celinski *et al.* and the theoretical calculations.[13] However, $0.6\mu_B$/Pd is nearly the same as the data given by Childress *et al.*[7] and Li *et al.* (see the inset in Fig. 3).[15] The source of the occurrence of the larger atomic moment of Pd in the sputtered Pd/Fe ML's may arise from the diffusion of Fe at the Fe-Pd interface.[7]

It is known that the Pd atoms adjacent to the Fe layers in the Pd/Fe system have a ferromagnetic polarization due to a strong hybridization between the d orbit of Fe and Pd. For the Pd atoms in the inner Pd layers, the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction may be used to describe their polarization behavior because the d orbit of Fe in the layers is not superimposed on the $4d$ orbit of Pd in the inner Pd layers. Such an indirect RKKY interaction makes the adjacent Fe layers coupled. The direct d-d interaction between Fe and Pd at the interface is stronger than the indirect RKKY interaction and in addition, the strength of the indirect one decays in an oscillating manner, with the spacer thickness, so that the adjacent Fe layers coupled ferromagnetically could be expected on the whole Pd thickness range. The change of the polarization of Pd from negative to positive, and vice versa, appears only in the space of the inner Pd layers, in other words, via modulating the Pd polarization in the inner Pd layers matches the ferromagnetic coupling between the adjacent Fe layers. Consequently, the interlayer coupling including both the direct and indirect interactions determine the magnetization oscillation in the Pd/Fe multilayers.

In the sputtered Pd/Fe ML's the diffusion of both Pd and Fe at the interface could be obtained more easily as compared with the samples prepared by MBE. The theoretical calculations usually assumed a sharp interface. Because the interface polarization, in turn, affects the polarization of Pd in the inner Pd layers, it is expected that there is a discrepancy between the results obtained by the different groups and between the experiment and the calculation. This point may be confirmed in the case of PdFe/Fe ML's. If a small amount of Fe impurity enters in the Pd layers, the long-range polarization of Pd/Fe ML's makes a change.

Figures 5(a)–5(c) show the M_s value averaged on the total Fe layers in $Pd_{1-x}Fe_x$/Fe ML's, as a function of PdFe layer thickness at $x=0.034$, 0.056, 0.071, respectively. Notice that the contribution from the buffer is already subtracted, although it is small. We found that both $Pd_{0.965}Fe_{0.034}$/Fe and $Pd_{0.944}Fe_{0.056}$/Fe ML's have a very similar oscillatory curve with the Pd/Fe ML's, i.e., having nearly the same oscillatory phase except an extra oscillatory period when $t_{Pd} \geqslant 12$ Å appears for $x=0.034$ and 0.0556, as compared with Pd/Fe ML's. The changes of the phase and the number of the oscillatory periods appear just when the density of Fe in Pd layer increases to 0.071. It means that the primary interactions existing in Pd/Fe ML's are prevailing to determine the oscillation phase when $x=0.034$ and 0.056 and the interaction between the impurities of Fe in the Pd layer may become dominant when $x=0.071$. The unchanged oscillation phase and period may show that the oscillation phenomenon in Pd/Fe is not affected by a small amount of Fe diffusion, but by interlayer coupling. If the oscillation results from diffusion, then the oscillation phase and period should be different for the multilayers with $x=0.034$ and 0.056, respectively. Moreover, if we plot the maximum M_s value as a function of Fe concentration in the Pd layers, $x=0.034$, 0.056, and 0.071, then we found that M_s decreases linearly with the decrease of x. Extrapolating this line to x

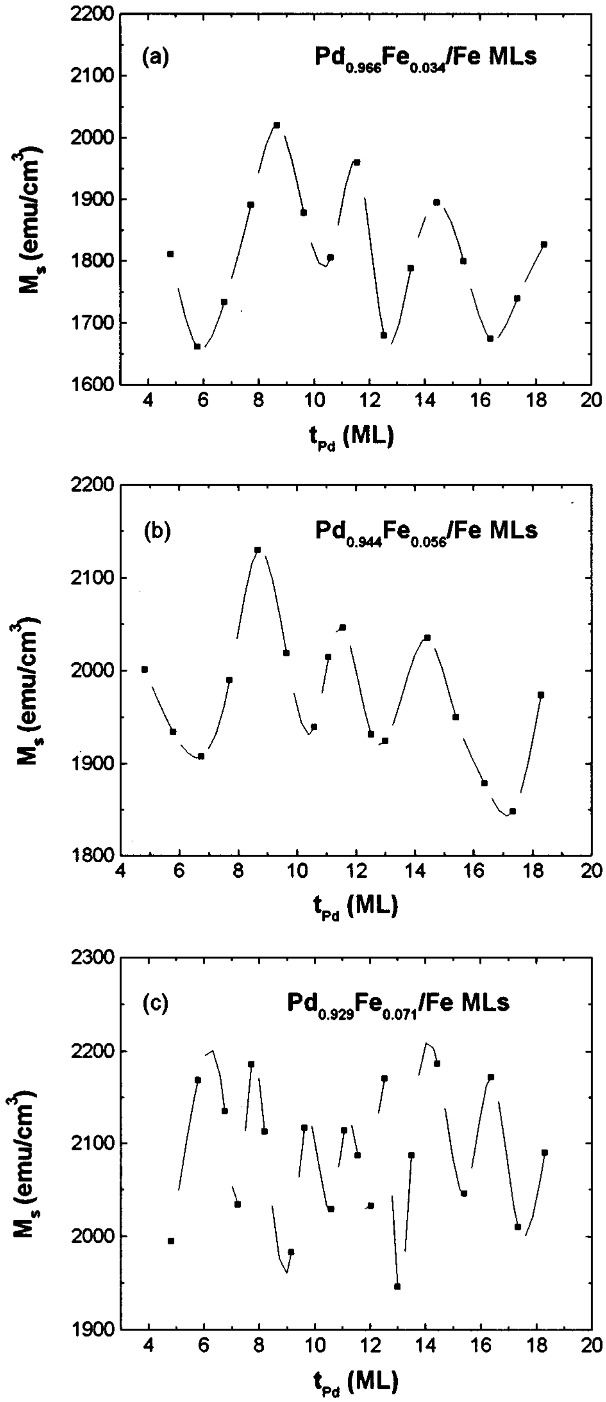

FIG. 5. Saturation magnetization M_s averaged on the Fe layers as the function of the Pd thickness t_{Pd} for the $Pd_{1-x}Fe_x$/Fe multilayers in the range of $x=0.034$, 0.056, and 0.071.

$=0$, we obtain $M_s \approx 1900\,\mathrm{emu/cm^3}$ that is the maximum value of M_s in the Pd/Fe multilayers. As a result, we speculate that Fe diffusion appears most probably at the interface region in Pd/Fe multilayers. It may be evidence of confirming that the long-range polarization of Pd in the Pd/Fe ML's is generated mainly by the interlayer coupling.

It can be seen from Fig. 5(a) that there are three valleys at $t_{Pd} \approx 6$, 12, and 16 ML on the curve of M_s vs t_{Pd} in the $Pd_{0.966}Fe_{0.034}$ multilayers. The respective M_s value is apparently smaller than that at the corresponding valleys in Pd/Fe ML's. It illustrates that nearly no extra magnetic moment in Pd layers is created at $t_{Pd} \approx 6$, 12, and 16 ML in $Pd_{0.966}Fe_{0.034}$/Fe multilayers. In other words, the existence of Fe impurity affects the spin configuration in the Pd layer. We also can see from Fig. 5 that M_s at the peaks where it shows a maximum value increases with the concentration of Fe in Pd layers as expected from the results in bulk magnetic dilute alloys.[16]

If we assume that the interactions between Fe layers through the PdFe layer and between the impurity of Fe atoms in the Pd layer are described by the RKKY interaction, then the total combined couplings in the PdFe/Fe ML's may be calculated, in principle, if the distribution of Fe impurities in the Pd layer is known. In other words, the spin configuration in the inner Pd spacer could be given and it may be determined by the spacer thickness and impurity density/distribution. Such work was done in the PtNi dilute alloy/Co ML's by Wang *et al.* recently.[17,18] According to their work, the total couplings in the PdFe/Fe ML's should be combined with the respective coupling between Fe in the layer and the impurity Fe in the inner spacer, and between the Fe impurities in the inner Pd layer. It is known that the phase in the RKKY interaction depends on the Fermi vector and it results from superposing all the RKKY contribution with the random phase occurring in the multilayers. Therefore, the resultant phase is determined by the impurity density/distribution. When the impurity density is low, the primary couplings in Pd/Fe prevail and then the oscillation phase remains in the $Pd_{1-x}Fe_x$/Fe ML's with $x=0.034$ and 0.056. When the concentration is increasing, it is possible for the resultant phase to change.

In PdFe/Fe ML's, Pd is polarized by both of Fe in the layer and the Fe impurities in the Pd layer, so that Pd having a larger atomic moment is expected. From the measurement, an averaged $\mu_{Pd} \approx 0.8\mu_B$ at $t_{Pd}=6$ ML is obtained in the $Pd_{0.929}Fe_{0.071}$/Fe multilayers and it is the maximum as compared with the $Pd_{1-x}Fe_x$/Fe ML's with $x=0.034$ and 0.056.

In conclusion, we have prepared the $Pd_{1-x}Fe_x$/Fe multilayers with $x=0$, 0.034, 0.056, and 0.071 by magnetron sputtering. It is confirmed from the measurement of the saturation magnetization in Pd/Fe ML's that M_s shows a long-range oscillation with a period of 4 ML thickness of Pd and that a negative polarization of Pd in the inner Pd spacer exists, which is different from the theoretical calculation. In the case of nearly ferromagnetic polarized Pd in the interface region, $\mu_{Pd} \approx 0.6\mu_B$. The larger value may arise from the interface diffusion. The phase and period of M_s oscillation remain after a small amount of Fe impurities, up to $x=0.056$, doping into the Pd layers as compared with the reference Pd/Fe multilayers. It may be concluded that the long-range oscillation of M_s in Pd/Fe ML's results from interlayer coupling rather than from Fe diffusion. Due to the entrance of Fe in the Pd layers, the saturation magnetization increases and it increases linearly with the Fe impurity density in the Pd layers.

ACKNOWLEDGMENTS

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