Effect of step decoration on the spin reorientation of Ni films grown on vicinal Cu(001)

U. Bauer,[1,2] J. Choi,[1] J. Wu,[1] H. Chen,[1,3] and Z. Q. Qiu[1]
[1]*Department of Physics, University of California at Berkeley, Berkeley, California 94720, USA*
[2]*Fakultät für Physik und Astronomie, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany*
[3]*Department of Physics, Tsinghua University, Beijing, People's Republic of China*
(Received 19 July 2007; revised manuscript received 18 October 2007; published 14 November 2007)

Ultrathin Ni films are grown on vicinal Cu(001) substrate with the steps parallel to the [110] axis and with the vicinal angle ranging from 0° to 12°. Spin reorientation transition (SRT) thickness of the Ni film is studied *in situ* using magneto-optic Kerr effect with step decoration of Mn, Fe, Co, Cu, and Pd. Surprisingly, the Ni SRT thickness is only affected by the Co step decoration with the other elements having little effect on the Ni SRT thickness. Further experiment using Co normal growth and a thin separation Cu layer confirms that the affected Ni SRT by the Co decoration comes from the Ni-Co interaction at the step edges.

DOI: 10.1103/PhysRevB.76.184415 PACS number(s): 75.70.Ak

I. INTRODUCTION

Magnetic crystalline anisotropy in magnetic crystals is generated by the spin-orbit interaction which transfers the spatial symmetry of the electron orbits to the electron spin freedom.[1] Magnetic anisotropy plays an important role in magnetic properties of materials, especially in low dimensional magnetic systems. For example, long-range magnetic order would not exist at any finite temperature in a two-dimensional Heisenberg system without the help of the magnetic anisotropy.[2,3] While the strength of the spin-orbit interaction depends sensitively on the electronic structure of the materials, the expression form of the magnetic anisotropy depends only on the crystal symmetry.[4] For example, uniaxial magnetic anisotropy, which assigns one easy/hard axis to the electron spin, could not exist in a three dimensional lattice with cubic symmetry but in a thin film where the translational symmetry is broken in the film's normal direction. For a thin film with in-plane square lattice [e.g., the (001) film of bcc or fcc lattice], in-plane uniaxial magnetic anisotropy cannot exist either due to the fourfold rotational symmetry. Because the overall magnetic anisotropy depends on both the spin-orbit interaction strength and the lattice symmetry-breaking, it is very difficult to trace the behavior of the magnetic anisotropy by simply comparing one system to the other system. In an attempt to separate the lattice symmetry-breaking from the spin-orbit interaction strength in the magnetic anisotropy, magnetic thin films grown on vicinal surfaces have been studied recently. It is found that the atomic steps on a (001) vicinal surface breaks the fourfold rotation symmetry to induce an in-plane uniaxial magnetic anisotropy.[5–7] This step-induced magnetic anisotropy has a profound effect on various properties of magnetic thin films such as the enhancement of the Curie temperature,[8] spin canting structure,[9] anisotropic susceptibility,[10] and the spin reorientation transition (SRT),[11] etc. To better understand the step-induced magnetic anisotropy, curved substrates were applied to explore the relation between the step-induced magnetic anisotropy and the step density,[12] and the result shows that different lattice symmetry seems to induce different forms of the step-induced magnetic anisotropy.[13] X-ray magnetic linear dichroism measurements confirm that spin-orbit interaction indeed changes at the step edges, supporting the fact that the step-induced magnetic anisotropy comes from the electronic change at the step edges.[14]

Although the above result indicates a localized effect of the step-induced magnetic anisotropy in the lateral direction, thickness-dependent study shows that the step-induced magnetic anisotropy does not have a simple one-over-thickness behavior,[15] showing a nonlocalized effect in the vertical dimension. To further understand the step-induced magnetic anisotropy, research on vicinal surfaces has been recently advanced by developing quasi-one-dimensional systems, either by growing magnetic wires on vicinal surfaces[16–18] or by step decoration of foreign atoms on a stepped magnetic thin film.[19] In the latter case, it is shown that side growth of foreign atoms can effectively decorate the step edges of a stepped film. The experiments, however, seem to show controversial result with the step-induced magnetic anisotropy depending sensitively on the step decoration for some systems[20] but insensitive for other systems.[15] Theoretical calculation using tight-bonding model suggests that the change of the step-induced magnetic anisotropy by step decoration comes from both the *d*-orbit change at the step edges and the structural relaxation near the step edges.[21] While the calculation could successfully explain the in-plane SRT of stepped Co film with Cu decoration, it is difficult to extend the calculation result to other systems which show a negative effect to the step decoration. Obviously, a systematic study of the step decoration by experiment is needed in order to provide a guideline for future theoretical calculation. In this paper, we studied the SRT of Ni film on curved Cu(001) with step decoration using the elements of Mn, Fe, Co, Cu, and Pd. Ni film grown on Cu(001) has a volume-type out-of-plane magnetic anisotropy and a surface-type in-plane magnetic anisotropy, leading to a SRT from in-plane to out-of-plane with increasing the film thickness.[22] Therefore, the addition of the step-induced magnetic anisotropy is expected to shift the Ni SRT thickness. Then a comparison of Ni SRT thickness with step decoration of different elements should provide some insight on the step-induced magnetic anisotropy. The five decorating elements we choose are around the element Ni in the Periodic Table and represent antiferromagnetic, ferromagnetic, diamagnetic, and paramagnetic elements, thus

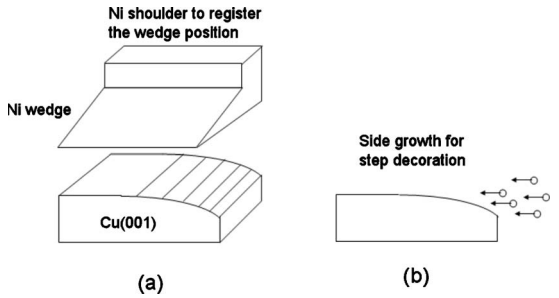

FIG. 1. Schematic drawing of (a) the Ni/curved Cu(001) system and (b) side growth for the step decoration.

should cover a wide spectrum of different electronic structures. Our experiment shows a surprising result that only the Co step decoration produces a significant effect on the Ni SRT thickness, while the other four elements have virtually no effect on the Ni SRT.

II. EXPERIMENT

A Cu(001) single crystal disk of 10 mm diameter and 2 mm thick was polished into a curved shape with the steps running along the [110] in-plane direction. The vicinal angle α varies continuously from 0° up to 12°. The substrate was cleaned *in situ* by cycles of Ar ion sputtering at 2 keV and annealing at 600 °C. All samples were grown at room temperature in vacuum with pressure below 9×10^{-10} torr. A water-cooled quartz balance was used to calibrate the evaporation rate prior to the sample growth. The Ni film was grown into a wedge shape (0–20 ML with 4 ML/mm) by moving the sample behind a knife edge shutter and with the wedge slope parallel to the substrate vicinal step edges. At the end of the wedge, a thick Ni shoulder of 40 ML was grown for the purpose of registering the Ni wedge position [Fig. 1(a)]. For the step decoration, the substrate was rotated 90° with the curved side of the substrate facing the deposition flux direction [Fig. 1(b)]. In this way, the deposited atoms are forced to reach the step edges and each step should receive the same amount of decoration atoms regardless of the vicinal angle. In this paper, we fix the step decoration amount to be one row of atoms per step for all of the five different elements so that a direct comparison can be made. Low energy electron diffraction (LEED) was used to characterize the sample. The sharp LEED spots before and after the film growth show that high quality single crystalline films have been achieved (Fig. 2). On the vicinal surface, the regular arranged atomic steps should split each LEED spot into double spots with their separation proportional to the vicinal angle.[6] Indeed we observed split LEED spots on our vicinal surface whose splitting increases with the vicinal angle. The slight elongation of the LEED spots is probably due to the finite size of the LEED electron beam which covers a range of the vicinal angle. Nevertheless, the split LEED spots indicate that well aligned atomic steps are formed before and after the Ni film growth. The sample was then transferred to the stage where the electromagnet resides for the magneto-optic Kerr effect (MOKE) measurement. The local vicinal

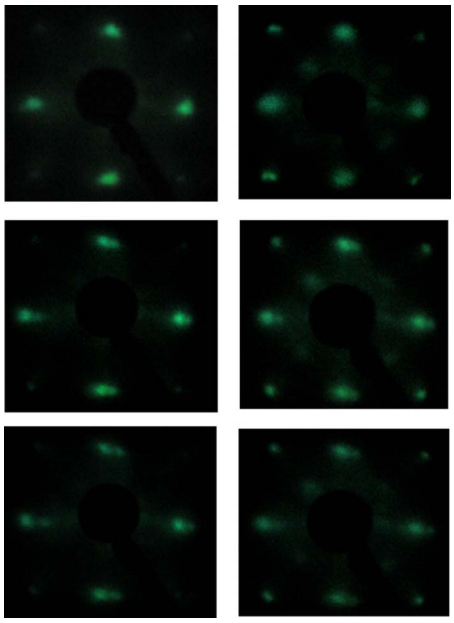

FIG. 2. (Color online) LEED diffraction patterns of 10 ML Ni (right column) on curved Cu(001) (left column) at $\alpha=0°$ (top), $\alpha=4°$ (middle), and $\alpha=8°$ (bottom), respectively. The electron energy is 132 eV.

angle was determined by the reflection angle of the MOKE laser beam. The finite beam width (~0.2–0.3 mm) covers a range of vicinal angle so that the reflection beam spreads into a range of ~1–2°. To reduce the error bar of the vicinal angle in the MOKE measurement, a slit was placed in the path of the reflected beam to narrow the vicinal angle spreading <0.2° in the MOKE measurement.

III. RESULT AND DISCUSSION

The Ni SRT was studied by taking the polar magnetic hysteresis loop (magnetic field perpendicular to the film) measurements at room temperature. Figure 3 shows the remanent polar Kerr signal, which is proportional to the perpendicular component of the magnetization $M_{\perp}$, as a function of the Ni film thickness. Representative polar Kerr loops at dif-

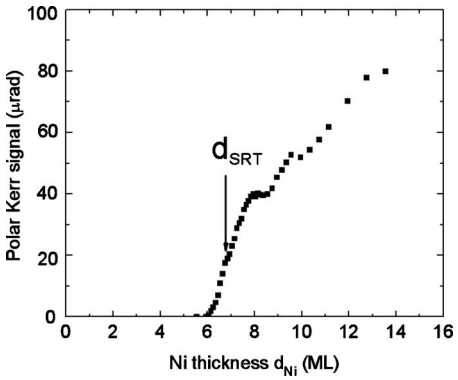

FIG. 3. Polar remanence of Ni film grown on flat Cu(001). Arrow indicates where the spin reorientation transition takes place.

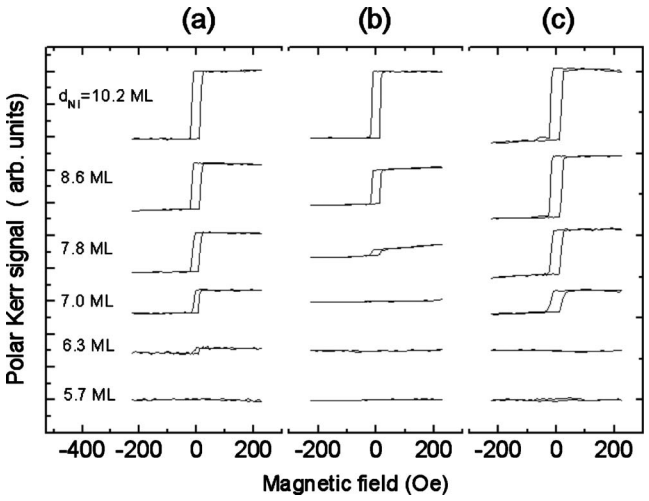

FIG. 4. Polar hysteresis loops of Ni film on vicinal Cu(001) with α=4° (a) pure Ni, (b) after Co step decoration using side growth, and (c) after normal growth of 0.07 ML Co.

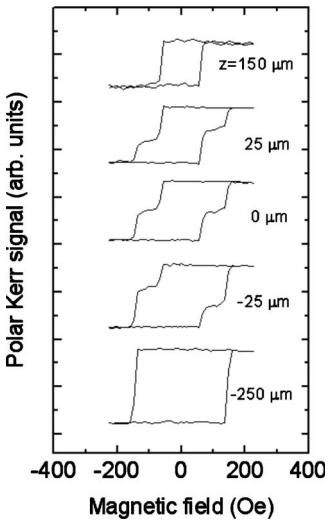

FIG. 5. Polar hysteresis loops near the Ni wedge/shoulder position. *Z* denotes the position of the Ni wedge. The steps in the hysteresis loop are used to register the Ni wedge position (see text).

ferent thicknesses are shown in Fig. 4(a). The rapid increase of the polar Kerr signal above 7 ML Ni signatures the Ni SRT from in-plane to out-of-plane directions at ~7 ML thickness. For practical convenience, we define the Ni SRT transition thickness d_{SRT} as that which the polar Kerr remanence reaches ~20 μrad (indicated by the arrow in Fig. 3). Then the Ni SRT transition thickness was determined as a function of the vicinal angle. For the pure Ni film, we find that the Ni SRT thickness has little dependence on the vicinal angle, suggesting that the atomic steps do not have a significant effect on the SRT thickness. Since the Ni wedge was grown at the same time on the flat and on the curved part of the substrate, the α dependence of d_{SRT} should be independent of growth rate variations. Here α and d_{SRT} are the vicinal angle and the SRT thickness, respectively. However, if the wedge position was not registered carefully, a wedge position shift at different vicinal angles could cause an artificial error in the determination of d_{SRT}. To ensure that we do not have this artificial error, we determined the Ni wedge position from the thick Ni shoulder at each vicinal angle. The 40 ML Ni shoulder has a greater coercivity than the 20 ML Ni film at the end of the wedge, thus a step in the Ni hysteresis loop due to these two different coercivities is expected once the MOKE laser beam covers the Ni wedge-shoulder boundary. This step should be exactly at the middle of the hysteresis loop when the center of the MOKE laser beam is exactly at the position of the wedge end (Fig. 5). Moving away from the shoulder position causes a change of the step position in the hysteresis loop. From Fig. 5, it is obvious that we can register the Ni position with an accuracy of ±25 μm in position. Given the Ni wedge slope of 4 ML/mm, a 25 μm position change is equivalent to a thickness change of 0.1 ML. Therefore we can claim that the d_{SRT} value determined in our experiment has an accuracy of ±0.1 ML.

Step decorations with Mn, Fe, Co, Cu, and Pd were performed on the Ni wedge with the amount of decoration atoms being 1 row/step for all the elements. The Ni d_{SRT} was then determined by MOKE measurement and the result is shown in Fig. 6. Surprisingly, only Co step decoration has a significant effect to increase the d_{SRT} while all other elements of Mn, Fe, Cu, and Pd have virtually no effect at all on the Ni SRT thickness. To ensure that the effect of Co on the d_{SRT} comes from the step decoration, we did two more experiments. First, we deposited 0.07 ML Co (~equivalent to 1 row/step at 4° of vicinal angle) onto the Ni wedge using normal growth instead of side growth. Co atoms on Ni surface do not have a high mobility to migrate to the Ni step edges as in the case of Cu on a Co surface[20] thus the Co atoms grown in this way stay mostly on the Ni surface rather than decorating the Ni step edges. MOKE measurements were then performed. For a comparison, MOKE hysteresis loops at different Ni thicknesses are shown in Fig. 4 for the Co side growth [Fig. 4(b)] and for the Co normal growth [Fig. 4(c)] at 4° of vicinal angle. While the Co step decoration with side growth obviously increases the Ni SRT thickness, the normal growth of Co has little effect on the d_{SRT}.

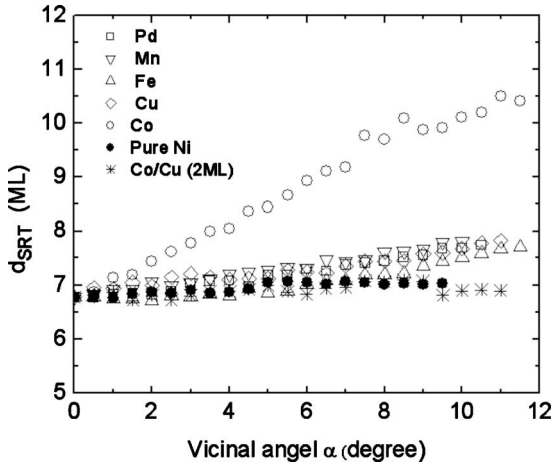

FIG. 6. Ni spin reorientation transition thickness vs the vicinal angle with step decoration of different elements.

This result shows that the effect of Co on the Ni d_{SRT} comes mainly from the Ni-Co interaction at the Ni step edges rather than from the Ni-Co interaction on top of the flat Ni surface. In the second experiment, we first grew 2 ML Cu to cover the Ni wedge and then performed the Co step decoration with side growth. In this way, the 2 ML Cu is expected to separate (or weaken) the Ni-Co interaction at the Ni step edges. Indeed MOKE measurement shows that the Co step decoration in this way has little effect on the Ni d_{SRT} (* in Fig. 6). The above two experiments prove that the effect of the Co step decoration on the Ni SRT comes from the Co-Ni interaction at the Ni step edges. This conclusion is further supported by the fact that the d_{SRT} increases monotonically with the vicinal angle for the Co decorated Ni film.

Then the interesting question is as follows: Why does only Co has a significant effect but the other four elements do not? We discuss several possibilities here. First, the addition of Co atoms is equivalent to the addition of a thin ferromagnetic film and thus should increase the in-plane magnetic shape anisotropy of $2\pi M^2$. This effect could be appreciable even if the amount of Co is small, because the magnetic moment of Co is about 3–4 times greater than the magnetic moment of Ni. But we rule out this possibility because the normal growth of Co on a Ni film does not affect the Ni SRT thickness. The next possibility is the formation of Co chains at the Ni step edges which could result in a magnetic shape anisotropy to favor an alignment of the magnetization parallel to the step edges. Then this additional in-plane shape anisotropy should shift the SRT to a thicker Ni film thickness. It seems this mechanism could explain the Co decoration result alone, but will not work when analyzing the step decoration result of other elements. For example, fcc Fe on Ni is ferromagnetic below 4 ML Fe thickness,[23] and Fe has a greater magnetic moment than Co. Thus if the shape anisotropy of the Co chains were responsible for the Ni SRT thickness shift, the Fe step decoration would have produced the same effect as Co. The fact that Fe step decoration has no effect on the Ni d_{SRT} rules out the mechanism of the Co-chain shape anisotropy. The next mechanism is the strain effect due to the step decoration. For Ni/Cu(001) system, the lattice mismatch between Ni and Cu causes a tetragonal distortion of the Ni film which generates a perpendicular volume magnetic anisotropy. Then a modification at the Ni step edges could in principle modify the lattice distortion to change the perpendicular magnetic anisotropy. However, the fact that the Ni SRT thickness changes very little with the vicinal angle suggests that the atomic steps on the vicinal Ni surface have only a limited effect on the perpendicular magnetic anisotropy. After step decoration, the foreign atoms may cause a local strain change to change the step-induced magnetic anisotropy.[21] This decoration-induced strain is expected to be proportional to the lattice mismatch between the foreign atoms and the Ni lattice even though a single row of foreign atoms at the step edges does not form a lattice. Among the five elements we choose, Co certainly has less lattice mismatch to Ni than some of the other elements [e.g., Mn and Pd (Ref. 24)]. Therefore, local strain effect induced by the step decoration does not seem to explain our experimental observation.

After excluding the above mechanisms, the plausible mechanism left is the electronic hybridization at the step edges. Generally speaking, heavy elements have a stronger spin-orbit interaction to generate a stronger magnetic anisotropy. That is why Co/Pd and Co/Pt multilayers consist of a strong magnetic anisotropy. In our experiment, however, Pd decoration has little effect on the Ni SRT so that the effect of step decoration is unlikely caused by the increased spin-orbit interaction strength by the foreign atoms. Another possibility is the magnetic properties of the decorating elements. In our experiment, Mn, fcc Fe (<4 ML), Co, Cu, and Pd are antiferromagnetic, ferromagnetic, ferromagnetic, diamagnetic, and paramagnetic, respectively. The experimental result shows that there is no apparent relation between the effect of the step decoration and the magnetic state of the decorating element. This fact suggests that the strong modification of the step-induced magnetic anisotropy by the Co step decoration is unlikely originated from the Co ferromagnetic nature. Theoretical calculation shows that the major effect of the step decoration comes from the crystal field splitting of the *d* orbits due to the foreign atoms.[21] If the theory is correct then our experimental result suggests that the Co step decoration causes a much greater crystal field splitting on the Ni atoms at the step edges than the other four elements. From the point of view of Néel's pair bonding model (although the model does not hold quantitatively for metals), our result suggests that a Ni-Co bond should generate a much stronger uniaxial magnetic anisotropy than the bond between Ni and other four elements. This mechanism needs further confirmation from future theoretical calculations. We here only provide our experimental fact in the hope of stimulating more theoretical interest.

IV. SUMMARY

In summary, we studied the SRT of Ni films grown on vicinal Cu(001) using step decorations with Mn, Fe, Co, Cu, and Pd atoms. We find that the Ni SRT thickness depends very little on the vicinal angle before the step decoration. After the step decoration, the Ni SRT thickness increases with the vicinal angle for Co step decoration but remains unchanged with the step decoration of the other four elements.

ACKNOWLEDGMENTS

This work was funded by NSF DMR-0405259 and ICQS of CAS. U.B. is supported by the Würzburg University Nanotechnology Exchange Program and by the Graduate Group in Nanoscale Science & Engineering at UC-Berkeley.

[1] D. S. Wang, R. Wu, and A. J. Freeman, Phys. Rev. Lett. **70**, 869 (1993).
[2] M. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966).
[3] M. Bander and D. L. Mills, Phys. Rev. B **38**, 12015 (1988).
[4] U. Gradmann, J. Korecki, and G. Waller, Appl. Phys. A: Solids Surf. **39**, 101 (1986).
[5] A. Berger, U. Linke, and H. P. Oepen, Phys. Rev. Lett. **68**, 839 (1992).
[6] J. Chen and J. L. Erskine, Phys. Rev. Lett. **68**, 1212 (1992).
[7] W. Weber, C. H. Back, A. Bischof, Ch. Würsch, and R. Allenspach, Phys. Rev. Lett. **76**, 1940 (1996).
[8] H. J. Choi, R. K. Kawakami, E. J. Escorcia-Aparicio, Z. Q. Qiu, J. Pearson, J. S. Jiang, D. Li, and S. D. Bader, Phys. Rev. Lett. **82**, 1947 (1999).
[9] S. S. Dhesi, H. A. Dürr, and G. van der Laan, Phys. Rev. B **59**, 8408 (1999).
[10] P. J. Jensen, S. Knappmann, W. Wulfhekel, and H. P. Oepen, Phys. Rev. B **67**, 184417 (2003).
[11] D. Repetto, T. Y. Lee, S. Rusponi, J. Honolka, K. Kuhnke, V. Sessi, U. Starke, H. Brune, P. Gambardella, C. Carbone, A. Enders, and K. Kern, Phys. Rev. B **74**, 054408 (2006).
[12] R. K. Kawakami, E. J. Escorcia-Aparicio, and Z. Q. Qiu, Phys. Rev. Lett. **77**, 2570 (1996).
[13] D. S. Chuang, C. A. Ballentine, and R. C. O'Handley, Phys. Rev. B **49**, 15084 (1994).
[14] S. S. Dhesi, G. van der Laan, E. Dudzik, and A. B. Shick, Phys. Rev. Lett. **87**, 067201 (2001).
[15] Y. Z. Wu, C. Won, and Z. Q. Qiu, Phys. Rev. B **65**, 184419 (2002).
[16] J. Shen, R. Skomski, M. Klaua, H. Jenniches, S. S. Manoharan, and J. Kirschner, Phys. Rev. B **56**, 2340 (1997).
[17] P. Gambardella, A. Dallmeyer, K. Maiti, M. C. Malagoli, W. Eberhardt, K. Kern, and C. Carbone, Nature (London) **416**, 301 (2002).
[18] R. Cheng, K. Y. Guslienko, F. Y. Fradin, J. E. Pearson, H. F. Ding, D. Li, and S. D. Bader, Phys. Rev. B **72**, 014409 (2005).
[19] R. Allenspach, A. Bischof, and U. Dürig, Surf. Sci. **381**, L573 (1997).
[20] R. K. Kawakami, M. O. Bowen, H. J. Choi, E. J. Escorcia-Aparicio, and Z. Q. Qiu, Phys. Rev. B **58**, R5924 (1998).
[21] M. Cinal and A. Umerski, Phys. Rev. B **73**, 184423 (2006).
[22] M. Farle, W. Platow, A. N. Anisimov, P. Poulopoulos, and K. Baberschke, Phys. Rev. B **56**, 5100 (1997).
[23] X. Liu and M. Wuttig, Phys. Rev. B **64**, 104408 (2001).
[24] It is difficult to obtain the structure when the decorating atoms form a single atomic chain. However, given the fact that atomic volume is difficult to change, it is safe to state that Co fits Ni better than Mn and Pd.