Angular effects of the critical current in Nb/Pd multilayers

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The angular dependencies of the critical current density, $J_c(\theta)$, for superconducting Nb/Pd multilayers which show, in parallel magnetic field, a pronounced peak effect, were investigated (here θ is defined as the angle from the plane parallel to the sample surface). We have demonstrated that the procedure of measurements is an important factor for the $J_c(\theta)$ shape. When rotating the sample in a fixed magnetic field H, it was found that the external field direction at which the maximum of J_c was obtained form an angle different from zero with respect to the planar direction. The angular position of the maximum depends on the external magnetic field, as well as on the initial conditions under which the samples were kept. It is found that the more intense the peak effect is on the $J_c(H)$ curves, the larger the angular shift is of $J_c(\theta)$ in these structures. The results are explained in terms of the magnetic field suppression of the percolation currents in a weak-link network. Flux trapped within grains by the flux pinning processes is relatively stable at low fields and its contributions to the transport current suppression add vectorially to the applied field.

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I. INTRODUCTION

One of the main sources of interest in superconducting artificial layered structures has been centered around the possibility to achieve higher critical currents.[1] The superconducting order parameter is strongly modulated in these structures. As a result, when the magnetic field is oriented along the layers, the centers of vortices are localized between superconducting films and a substantial energy barrier arises which prevents the vortices hopping between adjacent positions. After the discovery of high-T_c superconductors, which are usually natural layered materials, a lot of works appear devoted to the problem of experimental and theoretical study of vortex states in layered superconductors.[2] The vortex phase diagram is more complicated than that of conventional superconductors owing to small coherence lengths and large anisotropy. In spite of significant progress, which has been achieved in recent years by an understanding of the vortex states in layered superconductors with very weak Josephson coupling on the basis of the anisotropic London equations (see, for example, Refs. 3 and 4], some problems remain in this field.

The recent experimental developments of Nb-based layered structures, where insulating NbO layers played the role of effective pinning centers, demonstrated anomalous angular dependences of the critical current.[5] Later, a similar effect was observed on niobium films[6] and high-temperature superconducting tapes.[7] This phenomenon manifests itself in the fact that, under small external magnetic fields, the critical current attains its maximum in a slightly tilted field. This results from the difficulties of the transition of the flux vortex lattice into the ground state with minimum energy. Bringing the lattice into the ground state before every measurement at a new angle results in a symmetrical angular dependence of the critical current. On the other hand, it was found that the dependences of the critical current on the magnitude and on the direction of the external magnetic field change when varying the initial conditions under which these layered structures are kept.[5–7] In other words, by changing the initial position of a sample, one can vary the angular dependence of the critical current and even change the position of the maximum of the critical current of these structures by rotating them in a magnetic field. This fact allows us to control the critical parameters of superconductors during the experiment. It should be noted that the effect cannot be explained in the framework of modern theoretical works. Usually the planar pinning centers are believed[8] to be significantly weaker than the interlayer ones. As a result, the angular dependence of the critical current should have a maximum in parallel magnetic field.

Another interesting effect in the vortex properties of superconducting multilayers is the presence of peaks in the magnetic-field dependence of the critical current, first observed by Raffy *et al.* on Pb/PbBi proximity coupled multilayers.[9] Coccorese *et al.* observed this so-called peak effect also on Nb/Pd multilayers.[10] The first interpretation of this effect was given by Ami and Maki[11] in terms of commensurability between the multilayer period and the vortex lattice spacing. Pippard had previously explained the peak effect in superconductor as related to the adjustment of the vortex lattice with the pinning centers.[12] This approach was developed by Evetts and Glowacki[13] for polycrystalline superconductors.

To understand the nature of the phenomena observed in Refs. 5–7 and in Ref. 10 in greater detail, we carried out high-resolution angular measurements of the critical current in Nb/Pd multilayers. The choice of Pd is related to its large spin susceptibility. The Nb/Pd is an intermediate system between a well-known superconductor-normal metal system such as Nb/Cu and a superconductor-ferromagnet system. As a result, the properties of the multilayers are very sensitive to the Pd thickness.

II. EXPERIMENTAL

Three Nb/Pd multilayers with ten bilayers were grown on Si(100) substrates at room temperature by using a dual

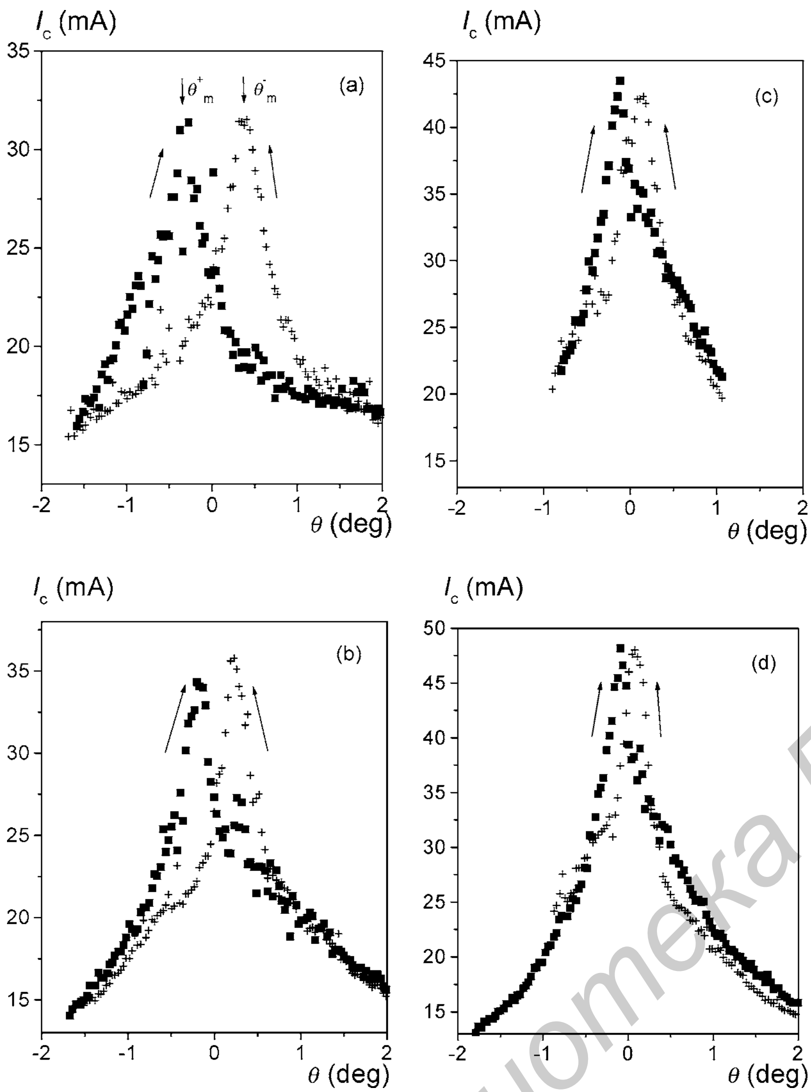

FIG. 1. Critical current through the sample with $d_{Nb}=20$ nm and $d_{Pd}=18$ nm as a function of the angle between the surface of the structure and the direction of the external magnetic field for various values of the field H. Squares correspond to the angular dependence recorded when the sample is rotated in the clockwise direction. Crosses represent the experimental results recorded when the sample is rotated in the counterclockwise direction. Arrows indicate the rotation direction: (a) $H=1.0$ kOe, starting angle of rotation is $-1.69°$; (b) $H=1.5$ kOe, starting angle of rotation is $-1.68°$; (c) $H=2.0$ kOe, starting angle of rotation is $-0.798°$; (d) $H=2.5$ kOe, starting angle of rotation is $-1.79°$.

source magnetically enhanced dc triode sputtering system as described earlier.[10,14] The samples had almost the same Nb thickness with variable Pd thickness ($d_{Nb}=20$ nm for $d_{Pd}=18$ nm and 20 nm, $d_{Nb}=18.7$ nm for $d_{Pd}=1.7$ nm). The samples were structured in shape of 10 μm wide bridges by photolithographic liftoff. The T_c values of patterned samples were around 6.3 K for the sample with $d_{Pd}=18$ nm and 20 nm and 7.0 K for sample with $d_{Pd}=1.7$ nm. Samples with different Nb thickness were prepared in different deposition runs. Structural properties of the Nb/Pd multilayer with $d_{Nb}=18.7$ nm were investigated in detail in Ref. 15, while for the samples with $d_{Nb}=20$ nm the results of structural investigations are similar to those reported in Ref. 14 for Nb/Pd multilayers having $d_{Nb}=25$ nm. The low-temperature resistivity value of Nb films was $\rho_{Nb}\approx 8\ldots 9$ $\mu\Omega$ cm. Comparing the superconducting properties of the multilayers with $d_{Pd}=18$ nm and 20 nm with those previously published,[10] we note that the critical temperature of Nb/Pd multilayers from Ref. 10 (where $d_{Nb}=18.7$ nm) is smaller, i.e., $T_c=3.65$ K for $d_{Pd}=17$ nm. Taking into account the similar values of ρ_{Nb}, the similar structural characteristics of both sets of the samples,[10,15] we may relate this discrepancy in T_c to the different d_{Nb} values. Indeed, as we showed recently, the T_c of Nb/Pd system, as well as the T_c of bare Nb films fabricated in our sputtering equipment, drop down very quickly for $d_{Nb}<20$ nm.[16]

The current-voltage characteristics were measured by the four-point probe technique. The critical current I_c was determined as the current at which the voltage drops on a sample reaches 1 μV. The detection of the critical current in our experiment was fully automated. This allowed us to determine the voltage drop with an accuracy of 0.1 μV. A magnetic field of intensity up to 65 kOe was produced by a superconducting solenoid. The $I_c(H)$ dependencies were measured at different temperatures for parallel orientation of H. The temperature stabilization was better than 10^{-3} K. For angular measurements, the sample was placed into the center of a solenoid onto a gear wheel, and could be rotated in such a way to vary the angle between H and the film plane while keeping the transport current always perpendicular to H. The sample can be rotated with respect to the direction of the external magnetic field by a precision worm-and-worm gear, made of stainless steel, with a ratio of 30. This fact provided an angular resolution of 0.04° for the sample orientation. All angular measurements were carried out at 4.2 K.

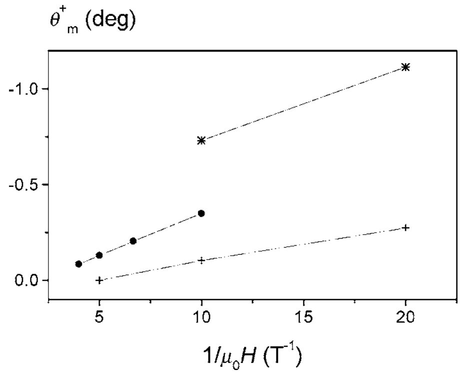

FIG. 2. The positions of the clockwise peak θ_m^+ as a function of H^{-1} for samples with d_{Pd}=20 nm (*), 18 nm (●), 1.7 nm (+), respectively.

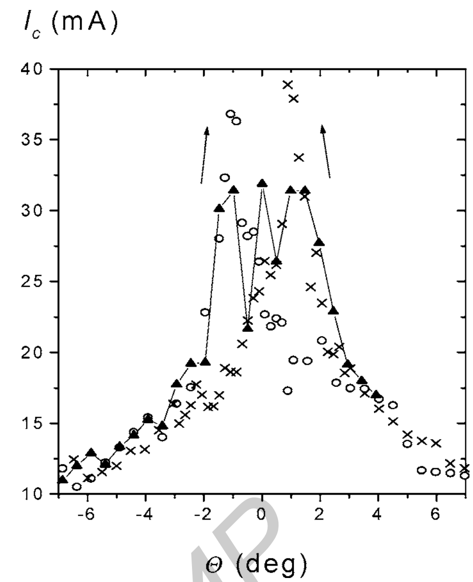

FIG. 3. The $I_c(\theta)$ dependences for the sample with d_{Nb}=20 nm and d_{Pd}=20 nm, H=0.5 kOe. Squares correspond to the angular dependence recorded when the sample is rotated in the clockwise direction. Crosses represent the experimental results recorded when the sample is rotated in the counterclockwise direction, and triangles show the results of normal-cycled experiment. The arrows indicate the rotation direction.

III. RESULTS

Figure 1 shows an example of the angular dependencies of the critical current for a sample with d_{Pd}=18 nm at four fixed magnetic field values, H=1.0, 1.5, 2.0, and 2.5 kOe. Here θ is the angle between the direction of the external magnetic field and the surface of the structure. The curves marked by closed squares correspond to the clockwise rotation (the positive direction), while the curves marked by crosses correspond to the rotation in the counterclockwise, or negative direction. For the clockwise rotations, the angle was varied from −30° to +30° for each value of the magnetic field, for the counterclockwise rotations, the angle was varied from +30° to −30°.

There are several noteworthy features in these dependencies. First, the peak in $I_c(\theta)$ is observed in the external magnetic field tilted with respect to the layers. Second, the positions of the clockwise (θ_m^+) and counterclockwise (θ_m^-) maxima do not coincide. Moreover, the values for θ_m^+ and θ_m^- are different for different values of the external magnetic field and the shift of the peak position from θ=0° increases when decreasing the external magnetic field. Figure 2 shows the positions of the clockwise peak as a function of H^{-1} for the three analyzed samples. As can be seen from the figure, the value of the shift diminishes with H enhancement and is approximately proportional to H^{-1}. In addition, the value of the shift is drastically different for the three samples: the shift increases with the increasing of Pd thickness. The position of the maximum is independent on the direction of the transport current. Thus, the positions of the clockwise and counterclockwise maxima depend on the initial conditions (memory effect) of a sample, as well as on the way in which it was brought to these conditions. *Third*, the value of the shift does not depend on the direction of the rotation and $\theta_m^-(H)=-\theta_m^+(H)$. These hysteresis dependencies have an important feature, i.e., the peak of $I_c(\theta)$ is observed before the orientation of the sample becomes parallel to the fixed magnetic field. It looks as if the response of the vortex system passes ahead of an influence on this system.

However, if a sample is brought into a normal state before each measurement of I_c at a new angle by increasing the magnetic field above the upper critical field, then these maxima will coincide and they will be observed when the external magnetic field is parallel to the plane of the multilayers (Fig. 3). In this case, the memory about the vortex structure for the previous angular position of the sample is absent. Evidently, the vortex lattice is less perturbed in this case than in a usual measurement when the external magnetic field is fixed. An example of the $J_c(\theta)$ dependence in the normal-cycled experiment is shown in Fig. 3 by triangles. The deflection of the dependence from symmetrical shape is negligible in this case. The maximum broadens and its top develops three local maxima. The positions of two of them are close to those for the direct and reverse rotation of the sample, and the central one is located when the field is parallel to the surface of the sample. These results are qualitatively analogous to the results obtained on Nb/NbO multilayer structures and high-temperature superconducting tapes.[5–7]

Figures 4(a) and 4(b) show examples of the magnetic field dependencies of the critical current density $J_c(H)$ at different temperatures. The $J_c(H)$ curves present the well-known peak effect in parallel field. It is interesting to point out that the height of the peak increases when increasing the Pd thickness. For a sample with d_{Pd}=1.7 nm, the peak effect was absent.[10]

In this work we found a relation between the presence of the peak effect in the $J_c(H)$ behavior and the position of $\theta_m(H)$ in Nb/Pd multilayers. It should be noted that, as supported by the temperature dependencies of the upper parallel critical magnetic field $H_{c2\parallel}(T)$, the proximity coupling between the superconducting layers decreases when increasing the Pd thickness.[14] In fact, the sample with d_{Pd}=20 nm shows the bidimensional behavior in the whole temperature

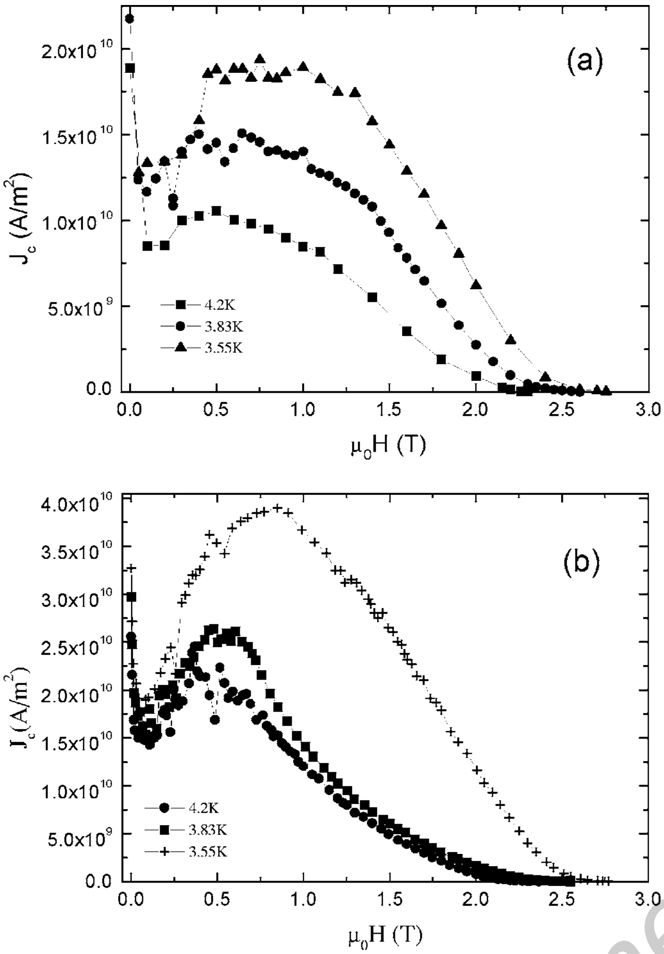

FIG. 4. The critical current density J_c vs parallel field at three temperatures (T=4.20 K, T=3.83 K, and T=3.55 K) for Nb/Pd multilayers with d_{Pd}=18 nm (a) and 20 nm (b), respectively.

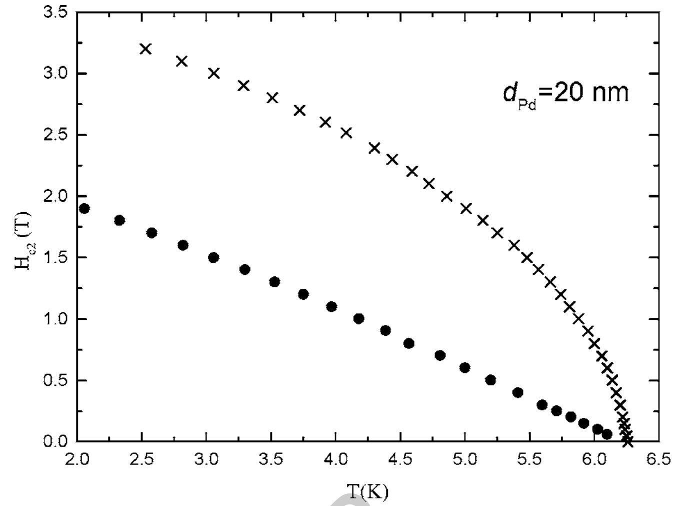

FIG. 5. The H_{c2} vs temperature dependence for multilayers with d_{Pd}=20 nm. Crosses correspond to the parallel magnetic field and dots to the perpendicular magnetic field.

range, up to T_c (Fig. 5). The sample with d_{Pd}=18 nm shows traces of the temperature change from linear to square-root-like behavior of $H_{c2\|}(T)$ [which usually is associated with the decoupling of the superconducting films in signal-to-noise ratio (S/N) multilayers] only very close to T_c. For the sample with d_{Pd}=1.7 nm, the change of linear $H_{c2\|}(T)$ dependence occurs well below T_c (see Fig. 2 of Ref. 6). The anisotropy values, $H_{c2\|}/H_{c2\perp}$ at T=4.2 K, were 2.8 for the samples with d_{Pd}=20 nm and 18 nm and 2.1 for the sample having d_{Pd}=1.7 nm. These are typical anisotropy values for proximity coupled multilayers, which reveals the two-dimensional (2D) behavior.[17]

We have to note that in Nb/Pd multilayers the interaction of the vortices with the planar defects (Pd layers) should give rise to higher critical current densities ($J_c \sim 10^{11}-10^{12}$ A/m^2),[1] but in our case this value is smaller ($J_c \sim 10^9-10^{10}$ A/m^2). Moreover, because the local maxima on the $J_c(H)$ curves depend on the matching of the vortex lattice to the layered structure, it should decrease when the interaction between the layers is weaker. The experimental results show an opposite behavior with the maxima in $J_c(H)$ for the sample with d_{Pd}=18 nm that are lower than the maxima for the sample with the weaker Josephson interaction (d_{Pd}=20 nm). Both these results indicate that are the processes in the Nb layers that define the critical current.[10] Taking into account the granular structure of our sputtered Nb films,[14] which is also confirmed by relatively large ρ_{Nb} values, it is reasonably to suppose that the lowered J_c values are due to the restriction caused by weak links between grains.

IV. DISCUSSION

An important point in our discussion is that the measured J_c values are lower with respect to the case in which they are determined by planar pinning centers because of the granular structure of sputtered Nb films in our multilayers. It is well known that in polycrystalline superconducting films the local intragrain critical current density, which can be measured, for example, by low-temperature scanning electron microscopy,[18] can exceed, by an order of magnitude, the global intergrain current density obtained by transport measurements.[19,20] That is why we believe that the approach based *only* on the commensurability between the period of the multilayer structure and the vortex lattice spacing cannot be used to clarify the nature of the peak effect. Moreover, this approach does not explain the crucial thickness dependence of the peak amplitude in the $J_c(H)$ curves. For these reasons, based on the observed correlation between the peak effect and the angular shift effect, we tried to find a model that might explain both these results at the same time. For the interpretation of the results, we used the hysteretic properties of a Josephson junction (weak link) network (normal or insulating barriers between Nb grains form weak links in polycrystalline films) controlled by the local magnetic field.[13] It is well known, in fact, that the critical current of such weak link is very sensitive to the magnetic field applied parallel to the barriers.[21] The key feature of the approach is that the local magnetic field is defined not only by the applied magnetic field but also the magnetic field of remanent trapped fluxes.

We believe that during the angle set measurements, we may always find an angle θ different from $\theta=0$, at which the diamagnetic properties of Nb, already for relatively small H values, are suppressed. Magnetic flux inside Nb can be trapped by the Nb grains. It means that, rotating the sample in the clockwise direction towards the parallel orientation of the external magnetic field, we reach the situation when the external magnetic field does not penetrate any more inside Nb layers. The resulting paramagnetic moment due to the previously trapped fluxes will drive a reverse flux component B_{rev} into Nb layers through the weakest junction

$$B_{\mathrm{rev}} = -kM_{\mathrm{tr}},$$

where k is a coefficient that is determined by the geometrical parameters of the layered structures and M_{tr} is the magnetic moment trapped by Nb grains. The M_{tr} value is determined by the pinning forces in Nb grains and by the applied field.

Because the B_{rev} is oriented in the opposite direction with respect to the perpendicular component of the applied magnetic field, the resulting fields in the weak link regions will be weaken. At angle θ_{m}^{+}, this cancellation reaches its maximum value, which leads to the maximum in $I_{\mathrm{c}}(\theta)$ dependence. At this angle

$$H \sin \theta_m^{+} = B_{\mathrm{rev}},$$

so that

$$\theta_m^{+} = -\sin^{-1}\left(\frac{kM_{\mathrm{tr}}}{H}\right). \tag{1}$$

When $H \gg kM_{\mathrm{tr}}$, Eq. (1) reduces to

$$\theta_m^{+} \approx -\frac{kM_{\mathrm{tr}}}{H}, \tag{2}$$

which explains the H^{-1} dependence of θ_{m}^{+} shown in Fig. 2. Moreover, this model points out that the peak in $I_{\mathrm{c}}(\theta)$ should be observed *before* the sample reaches the parallel orientation, in very good agreement with the experimental data.

In the region $\theta_{\mathrm{m}}^{+} < \theta < 0$ the magnetic field of the trapped fluxes becomes larger than the perpendicular component of the external field and I_{c} diminishes. In the parallel field, just the remanent trapped fluxes define the critical current of weak links. Moreover, due to the pinning a few paramagnetic moments could exist even in the case of the opposite direction of the perpendicular component of the external field. Nevertheless, at some angle, the diamagnetic properties of the Nb layers will be suppressed again, and the external field penetrates into Nb granules and in weak links like in the initial stage. In this case, a maximum level of cancellation between the applied field and reverse flux due to the remaining trapped flux is achieved also *before* the orientation of the sample becomes parallel to the external magnetic field. Evidently, the positions of the clockwise and counterclockwise maxima do not coincide, in agreement with the experiment.

In the considered picture we did not mention the possible perpendicular component of the magnetic field generated by the transport current. This component can be vectorially added to the trapped flux to give the correct values of θ_{m}^{+} and θ_{m}^{-}. In any case, taking into account this additional component of the magnetic field does not change the main points of our results. However, when we analyze the $J_{\mathrm{c}}(H)$ measurements in external parallel magnetic field, the perpendicular component of the magnetic field is the field generated by the transport current. Evidently the trapped flux should disappear with increasing of the external field, because this field forces the paramagnetic moments to draw up along its direction. As a result the intergrain critical current increases giving a local maximum in the $J_{\mathrm{c}}(H)$ dependencies. Therefore, in our picture, the peak effect is related to the adjustment of the vortex system to the granular structure.

It is worth mentioning that the dependence of the $J_{\mathrm{c}}(H)$ peak effect and of the effect of the angular shift by the small increase of Pd thickness could also be explained by considering the high paramagnetic nature of Pd,[16,22] which should increase the reverse flux component directed into Nb layers through the weakest junction. With increasing Pd thickness, this effect is enhanced and this, in turn, increases the peak effect. At this point, the cancellation between the applied field and the reverse flux due to the remaining trapped flux can be reached in Nb/Pd system at greater θ values for thicker Pd layers, in agreement with the experiment (Fig. 2).

V. CONCLUSIONS

Anomalous angular dependencies of the critical current in superconducting Nb/Pd multilayers have been observed. This anomaly manifests itself in the fact that, for small intensity of the external magnetic field, the critical current attains its maximum in a slightly tilted magnetic field, and it is associated with the complex character of the transition of a vortex system to the unperturbed ground state. In standard critical current measurements, the vortex system is in an excited metastable state but their interpretations are based on theoretical studies that deal with the equilibrium properties of the vortex system in superconductors. In this paper we have shown that both the peak effect on the $J_{\mathrm{c}}(H)$ curves and the angular shift on the $J_{\mathrm{c}}(\theta)$ dependences in Nb/Pd multilayers can be explained in terms of the magnetic field suppression of percolation currents in a weak link network due to the granularity of Nb layers.

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