

# Critical fields and dimensional crossover in layered periodic and nonperiodic superconductors

N. Ya. Fogel' and V. G. Cherkasova

*Low-Temperature Physicotechnical Institute, 310164 Khar'kov, Ukraine*

(Submitted 6 June 1995)

*Zh. Éksp. Teor. Fiz.* **109**, 223–237 (January 1996)

In this paper we present the results of our experimental studies of critical magnetic fields for two types of layered superconductors—types S/I and S/N—as well as S/I/S sandwiches. A characteristic feature of the behavior of both layered and sandwich systems is dimensional crossover, whose presence determines both the temperature dependence of the parallel critical field  $H_{c\parallel}$  and the magnetic field dependence of the derivative of the critical field with respect to angle near the parallel orientation  $\beta = dH_{c2}/d\theta|_{\theta=0^\circ}$ . We discuss various types of crossover ( $3D-2D$ ,  $2D'-2D$ ,  $2D-3D$ ), and consider the mechanisms that determine the change in dimensionality in layered superconductors. We establish that all types of crossover can be observed in both superlattices and sandwiches, which may be regarded as the elementary blocks from which the superlattices are constructed, when the layer parameters are appropriately chosen. For both superlattices and sandwiches, we find that in all cases the functions  $H_{c\parallel}(T)$  are essentially indistinguishable, and that only by measuring  $\beta(H)$  can we establish the detailed nature of the nonuniformity of a sample in the direction normal to the layers and the degree of ideality of its macrostructure. © 1996 American Institute of Physics. [S1063-7761(96)01501-9]

## 1. INTRODUCTION

One of the most interesting phenomena observed in various superconducting layered periodic structures (e.g., natural layered compounds of the intercalated transition metal dichalcogenide type, high-temperature metal-oxide compounds, and artificial superlattices) is dimensional crossover, which affects both the temperature dependence of the parallel critical field  $H_{c\parallel}(T)$  and the fluctuation conductivity. In the majority of cases, crossover is brought about by changing the ratio between the coherence length  $\xi(T)$ , which determines the size of the superconducting seed, and the period of the layered structure  $D$ . Near the superconducting transition temperature  $T_c$  (both above and below it) the coherence length  $\xi(T) \sim (1 - T/T_c)^{-1/2}$  can be much larger than this period, and then the multilayered system behaves like a three-dimensional ( $3D$ ) anisotropic superconductor. At temperatures far from  $T_c$  this ratio can decrease until two-dimensional ( $2D$ ) behavior is observed.

In this paper, we will restrict our discussion to the behavior of the critical magnetic fields of layered superconductors in the presence of dimensional crossover. The type of crossover most often observed in layered systems is the  $3D-2D$  type, evidence for which is the replacement of a linear temperature dependence of the critical field  $H_{c\parallel}$  near  $T_{c\parallel}$ , i.e.,  $H_{c\parallel} \sim (1 - T/T_c)$  (the range of  $3D$  behavior), by square-root behavior, i.e.,  $H_{c\parallel} \sim (1 - T/T_c)^{1/2}$  at low temperatures (the range of  $2D$  behavior). This type of crossover is observed<sup>1</sup> in the intercalated compound  $\text{TaS}_2$  ( $\text{Py}_{1/2}$ ) and in a number of superconducting superlattices: in the S/N (superconductor-normal metal) type superlattices<sup>2,3</sup> Nb/Cu, V/Ag, the S/S' (superconductor–superconductor) type<sup>4,5</sup> superlattices Nb/NbTi, Nb/Ta, and in the S/I (superconductor–

semiconductor) type superlattices Nb/Ge (Ref. 6), Pb/Ge (Ref. 7), V/Si (Ref. 8). A theoretical description of the crossover phenomenon for S/N systems in which the coupling between layers comes about through the proximity effect was proposed in Refs. 9 and 10, and a theory for multilayer periodic systems of S/I type, where Josephson coupling exists between the layers, was discussed in Refs. 8, 10, and 11.

Crossover is also observed in simpler layered structures of the sandwich type (i.e., two superconducting films separated by a layer of semiconductor).<sup>7,12,13</sup> In this paper we want to focus attention on the fact that in certain cases when crossover is observed the functions  $H_{c\parallel}(T)$  for sandwiches are outwardly indistinguishable from those for superconducting superlattices. Therefore, observation of a change in the character of the temperature dependence of the critical field in layered superconductors does not in itself indicate the presence of a superstructure or a high-quality superlattice.

More trustworthy evidence of the presence of regular periodic nonuniformity in the direction perpendicular to the layers and of the destruction of a superstructure can be obtained from measurements of the angular dependence of  $H_{c2}$ . This follows from the experimental data obtained for the superlattices<sup>14,15</sup> V/Si and Nb/Si from measuring the functions  $H_{c2}(\theta)$  and the magnetic field dependence of the derivative of the critical field on angle near the parallel orientation, i.e.,  $\beta = dH_{c2}/d\theta|_{\theta=0^\circ}$ , and also from the theoretical results of Ref. 16.

The task of this paper will be to investigate the parallel critical fields and the functions  $\beta(H)$  for various types of layered superconductor systems, and to compare the properties of regular multilayer systems with those of sandwiches.

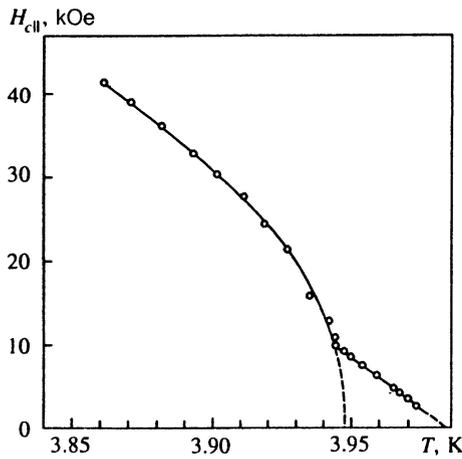


FIG. 1. Temperature dependence of the parallel critical field  $H_{c||}$  for a Mo/Si superlattice ( $D=55 \text{ \AA}$ ,  $d_{Si}=25 \text{ \AA}$ ,  $T_c=3.98 \text{ K}$ ).

## 2. METHODS FOR OBTAINING SAMPLES AND EXPERIMENTAL METHODOLOGY

Our layered composites were obtained by successively depositing material from two electron-beam evaporators in a vacuum chamber under a residual gas pressure of  $1.3 \cdot 10^{-6}$  to  $2.7 \cdot 10^{-7}$  Torr. The layer thicknesses were monitored by a quartz resonator, and the value of the superlattice period was refined based on small-angle x-ray diffraction. The materials were sputtered through special thermally stable masks with preset geometries, allowing us to use the four-probe method to investigate the electrical characteristics of the sputtered samples. The superconducting materials we used were films of the transition-metals Nb, Ta, V, and Mo; in the majority of our S/I type samples, we used a layer of silicon with thickness  $\sim 30 \text{ \AA}$  as the insulating layer. Whenever the thickness of the Si interlayer used differs from this value, we have noted the fact in footnotes to the figures. Our measurements of the electrical characteristics were made in a cryostat with a superconducting solenoid in the temperature range from 4.2 down to 1.6 K. While the measurements were being made, the samples were positioned at the center of the solenoid; a special rod with a rotating apparatus allowed us to vary the orientation of the sample with respect to the applied magnetic field. The error in determining the angle  $\theta$  between the directions of the magnetic field and the plane of the layers did not exceed  $0.1^\circ$ , since we used Hall pickups.<sup>15</sup> The temperature at the chosen point was stabilized to 0.003 K, and the magnetic field to 0.5% of its nominal value.

## 3. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows a typical temperature dependence of the parallel critical field  $H_{c||}$  of a superlattice that demonstrates  $3D-2D$  dimensional crossover. These data were obtained for a Mo/Si superlattice, i.e., a superlattice of S/I type. Analogous behavior of  $H_{c||}(T)$  is observed for superlattices of types S/S' and S/N as well.<sup>2,5</sup> For this superlattice, as for other superlattices of S/I type, all the parameters can be obtained by using the expressions of Refs. 8, 10,

$$H_{c||}(T) = \frac{\phi_0}{2\pi\xi^2(T)} \frac{\sqrt{ld}}{D} = \frac{\phi_0}{2\pi\xi^2(T)} \sqrt{\frac{M}{m}}, \quad H < H_{cr}, \quad (1)$$

$$H_{c||}(T) = \frac{\sqrt{3}\phi_0}{\pi d\xi(T)} \left(1 + \frac{2}{15} \frac{d}{l}\right)^{1/2} \left(\frac{\tilde{T}_c - T}{T_c}\right)^{1/2}, \quad H > H_{cr}, \quad (2)$$

$$\tilde{T}_c = T_c \left(1 - \frac{2\xi^2}{ld}\right), \quad (3)$$

$$H_{cr} = \frac{\phi_0}{\pi D \sqrt{ld}} = \frac{\phi_0}{\pi D^2} \sqrt{\frac{m}{M}}, \quad (4)$$

by measuring the temperature dependence of the parallel and perpendicular critical fields. Equation (1) describes the behavior of  $H_{c||}$  in the  $3D$  range, Eq. (2) in the  $2D$  range. Here  $D=d+s$  is the superlattice period,  $d$  is the thickness of the metallic layers,  $s$  that of the semiconductor interlayer,  $\xi(T)$  is the coherence length,  $\xi = \xi(T=0)$ ,  $(M/m)^{1/2} = (dH_{c||}/dT)/(dH_{c\perp}/dT)$  is the anisotropy parameter,  $H_{cr}$  is the crossover field,  $l$  is an extrapolation length determined by the boundary conditions at the junction between the metal and semiconductor layers that characterizes the strength of the coupling between layers, and  $\tilde{T}_c$  is an extrapolation temperature obtained by extrapolating Eq. (2) to  $H=0$ . The temperature  $\tilde{T}_c$  is always smaller than the superconducting transition temperature  $T_c$  of the superlattice as a whole, due to a distinctive proximity effect<sup>8</sup> (a layer containing a superconducting seed is in contact through thin insulating interlayers with metallic layers in which the superconductivity is suppressed by a magnetic field).

For the Mo/Si superlattice, the data for which are presented in Fig. 1, the fundamental parameters determined with the help of Eqs. (1)–(4) have the following values:  $T_c=3.95 \text{ K}$ ;  $\xi(0)=63 \text{ \AA}$ ;  $(M/m)^{1/2}=12$ ;  $H_{cr}=9 \text{ kOe}$ ;  $l=1.4 \cdot 10^{-4} \text{ cm}$ .

In superlattices, types of crossover are possible other than crossover from three-dimensional behavior near  $T_c$ , when the superconducting seed overlaps many layers of the superlattice, or from two-dimensional behavior at low temperatures, when the seed is localized at a single superconducting layer.<sup>8,14,17,18</sup> In particular, if the superlattice contains only a finite number of layers  $N$ , two-dimensional behavior is observed in the immediate vicinity of  $T_c$ , which is replaced by three-dimensional behavior as the temperature decreases (Fig. 2).

In Fig. 2 we show data for a V/Ta superlattice with period number  $N=5$ . This figure illustrates a sequence of characteristic temperature dependences of  $H_{c||}$  that is peculiar to this type of crossover:  $H_{c||} \sim (1-T/T_c)^{1/2}$  near  $T_c$ , and  $H_{c||} \sim (1-T/T_c)$  at lower temperatures. It is possible to choose the parameters of the superstructure and the number of layers in such a way that crossovers of  $2D'-3D$  and  $3D-2D$  type can be observed in the function  $H_{c||}(T)$  at the same time.<sup>17</sup> Like the  $3D-2D$  crossover, the  $2D'-3D$  crossover is produced by a change in the ratio between the size of the superconducting seed and the thickness; now, however, it is not the thickness of an individual superconducting layer, but rather the total sample thickness  $L$  that

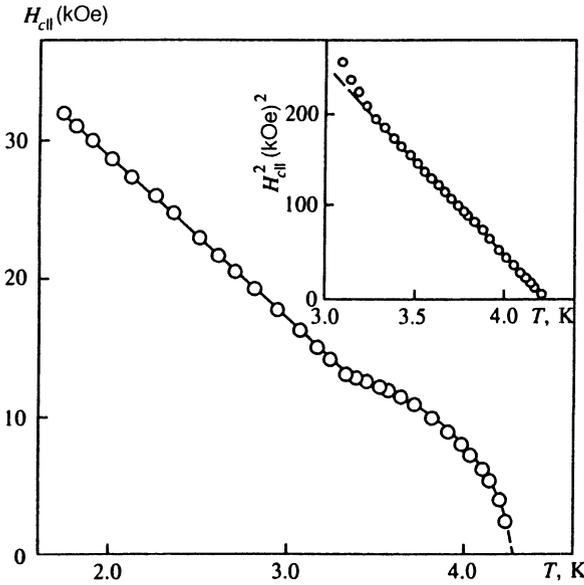


FIG. 2. Temperature dependence of the parallel critical field  $H_{c||}$  for a V/Ta superlattice ( $D=30$  Å,  $d_{Si}=30$  Å). In the inset we show the function  $H_{c||}^2(T)$  for temperatures close to  $T_c$ .

enters into the ratio. For  $L \gg \xi(T)$ , the superlattice behaves like a two-dimensional layer with thickness  $L \sim ND$ , while for lower temperatures a transition takes place to a one-dimensional mixed state, which is replaced by the ordinary mixed state at still lower temperatures. For uniform thin films, the parallel critical field<sup>19</sup> near  $T_c$ ,

$$H_{c||} = \frac{\sqrt{12} \phi_0}{2\pi L \xi(T)} \quad (5)$$

is proportional to  $(1 - T/T_c)^{1/2}$ . From experiments (Fig. 2 and the data of Ref. 17 on PbTe/PbS superconducting superlattices) it follows that the presence of a periodic nonuniformity in the superlattice does not affect the form of the function  $H_{c||}(T)$  at temperatures close to  $T_c$ . The value of  $L$  for a superlattice with a small number of layers calculated using Eq. (5) is in rather good agreement with its real value. Thus, from the data shown in Fig. 2 we obtain  $L=334$  Å for a nominal thickness value of 300 Å. The position of the  $2D' - 3D$  crossover temperature  $T_0$  at which the sample enters the one-dimensional mixed state, and which for the case of uniform films is determined by the relation<sup>20</sup>

$$L = \sqrt{\frac{5}{2}} \xi(T_0), \quad (6)$$

is in somewhat worse agreement with what is expected from the theory of uniform films. For the same V/Ta sample, the experimental value of the crossover temperature  $T_0 = 3.2$  K, whereas the value of  $T_0$  calculated from Eq. (6) is 2.96 K.

For superlattices of  $S/S'$  type [e.g., Nb/NbTi (Ref. 4), Nb/Ta (Ref. 5), and Nb/NbZr (Refs. 21 and 22)] a  $2D - 2D''$  type of crossover is also observed, connected with the change in the character of the two-dimensional behavior. This type of crossover, which is produced by the transfer of the superconducting seed from the  $S$ -layer to the  $S'$ -layer, was first predicted theoretically by Takahashi and Tachiki<sup>18</sup>

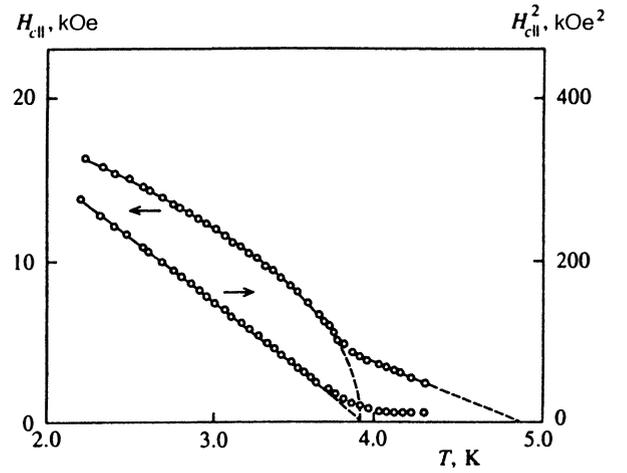


FIG. 3. Temperature dependence of the parallel critical field  $H_{c||}$  and the quantity  $H_{c||}^2(T)$  for an asymmetric V/Si/V sandwich consisting of thick ( $d_1=2000$  Å) and thin ( $d_2=500$  Å) metallic layers of vanadium.

for the case of two superconductors with close values of  $T_c$  and significantly different electron mean free paths, and then was observed experimentally.<sup>4,5,21,22</sup> The temperature of this crossover is always below  $T_c$  for the  $3D - 2D$  dimensional transition.<sup>5,21,22</sup>

Now let us turn to our results for various types of sandwich structures with semiconducting interlayers ( $S/I/S$ ), and compare them with data for the superlattices. We investigated both symmetric and asymmetric sandwiches. By “symmetric” sandwiches we mean layered systems consisting of two completely identical superconducting films separated by a tunneling interlayer. We call a sample asymmetric when its two metallic layers have different values of some characteristic parameter or set of characteristic parameters (e.g., critical temperature, thickness, mean free path, and accordingly coherence length at temperature  $T=0$ ).

First let us consider the data for asymmetric sandwiches. In Fig. 3 we show the temperature dependence of the parallel critical field of a sandwich consisting of a thick and a thin metallic layer ( $d_1=2000$  Å,  $d_2=500$  Å). In the same figure, we show the temperature dependence of the function  $H_{c||}^2$  for this sample. In layer 1, the superconducting transition temperature  $T_{c1}$  is higher than in layer 2. Near the superconducting transition temperature of the sandwich, which is essentially identical to  $T_{c1}$ , our experiments show that the parallel critical field is determined by the critical field for surface superconductivity  $H_{c3}$  of the thick ( $d \gg \xi(T)$ ) layer 1. At a certain temperature  $T^*$  a transition takes place to square-root behavior  $H_{c||}(T) \sim (\bar{T}_c - T)^{1/2}$ , which is characteristic of the parallel critical field of the thin layer 2 which for  $T^* < T_{c2}$  turns out to be larger than the critical field of layer 1. As follows from Fig. 3, the resulting dependence  $H_{c||}(T)$  turns out to be qualitatively the same as that observed in superlattices (see Fig. 1) for a  $3D - 2D$  crossover, although this layered sample contains only two superconducting layers.

The temperature dependence of  $H_{c||}$  for an asymmetric sandwich with two thin metallic layers is shown in Fig. 4. For this sample we observed an abrupt transition from one square-root dependence for  $H_{c||}(T)$  near  $T_c$  to another

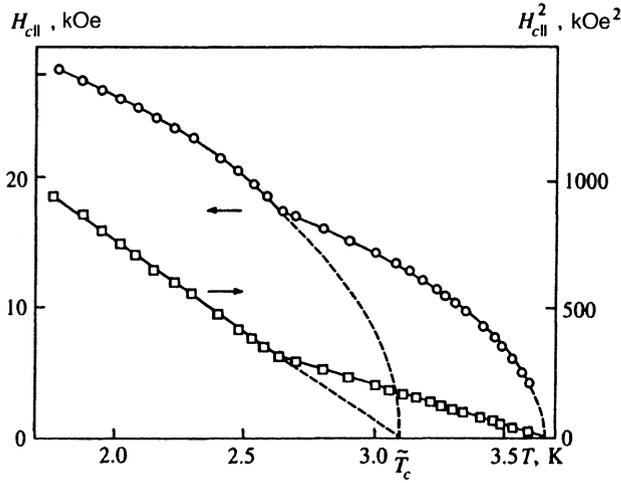


FIG. 4. Temperature dependence of the parallel critical field  $H_{c||}$  and the quantity  $H_{c||}^2(T)$  for an asymmetric thin-layer V/Si/V sandwich ( $d_1=200 \text{ \AA}$ ,  $d_2=300 \text{ \AA}$ ).

square-root dependence at low temperatures. This function demonstrates a  $2D'-2D$  type of crossover which corresponds to a transition between two different two-dimensional states.

The data for a symmetric sandwich made of thin vanadium layers is shown in Fig. 5. As follows from the figure,  $2D'-2D$  crossover is also observed in such sandwiches. An analogous form of crossover is observed in symmetric thin-layer Pb/Ge sandwiches as well, for germanium thicknesses of 20 and 25  $\text{\AA}$ .<sup>7</sup> It is clear from Figs. 4 and 5 that the functions  $H_{c||}(T)$  for thin-layer symmetric and asymmetric sandwiches are qualitatively similar; however, there are also certain important differences. For the symmetric sandwich, the transition from one square-root dependence to another is

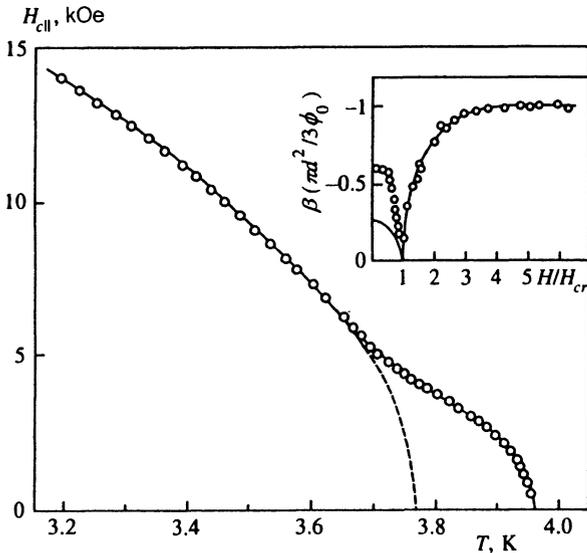


FIG. 5. Temperature dependence of the parallel critical field  $H_{c||}$  for a symmetric thin-layer V/Si sandwich ( $d_1=d_2=300 \text{ \AA}$ ). In the inset, we give the dependence of the normalized derivative of the critical field with respect to angle for  $\theta=0^\circ$  on the reduced field  $H/H_{cr}$  for the same sample. The solid curve is the theoretical dependence calculated using Eqs. (10) and (13).

smooth, whereas for the asymmetric sandwich this transition is discontinuous in character.<sup>12,13</sup> For symmetric sandwiches the slope of the function  $H_{c||}(T)$  at crossover always changes by a factor of 4, independent of the thickness of the metallic layers (if  $d \ll \xi(T)$ ), whereas for asymmetric thin-layer sandwiches the ratio of the slopes of  $dH_{c||}^2/dT$  at temperatures higher than and lower than the crossover point depends on the ratio between the thicknesses  $d_1$  and  $d_2$ , and also on the values of  $\xi_1$  and  $\xi_2$  in each of the layers.<sup>12,13</sup>

Thus, the experimental data presented here imply that exactly the same dependence  $H_{c||}(T)$  is observed for a sandwich as for  $3D-2D$  and  $2D-2D''$  dimensional crossovers in superlattices. For superlattices, it is customary to assume that a change in the character of the temperature dependence of  $H_{c||}(T)$  corresponding to  $3D-2D$  crossover is the most typical manifestation of discreteness and regularity of the superlattice structure. However, a comparison of the superlattice results with data obtained for a sandwich indicates that the simple fact that the temperature dependence of the parallel critical field changes abruptly as the temperature falls is insufficient to identify superlattice effects. In cases where processing in preparing the superlattice is imperfect, various disruptions of the ideal periodicity of the superlattice are possible, in particular "fusing" of the metallic layers due to shorting across the semiconductor interlayers. It is clear from the results presented above (Fig. 3) that only one weak link in the sample is enough to let us observe effects similar to  $3D-2D$  dimensional crossover in superlattices. Thus, in order to verify the presence of the periodic discrete structure of the superlattice we require more refined experiments than simply measuring the function  $H_{c||}(T)$ . These experiments will be the subject of the discussion that follows.

On the other hand, it is not difficult to show that "standard" superlattice effects can be obtained even for two identical superconducting layers with different thicknesses separated by a thin insulating interlayer. It is clear that near the transition temperature, where the energy of a superconducting seed  $e^2 H^2 d^2 / mc^2$  (calculated for a pair of superconducting electrons) is smaller than the corresponding weak-coupling energy between layers  $\hbar^2 / mld$ , the superconducting order parameter in these samples is different from zero in both films, and is essentially constant over the sandwich thickness. In this case, the parallel critical field of the sandwich equals the critical field of a film with twice the thickness (for  $s \ll d$ ). As the temperature falls, the "magnetic" energy increases, and then it becomes advantageous to localize the seed in one of the layers. In this case, naturally, the slope  $dH_{c||}^2/dT$  should increase by a factor of 4.<sup>12</sup> Just this change in the slope is observed for all the symmetric sandwiches we investigated. Localization of the order parameter in one of the layers takes place in fields<sup>12</sup>

$$H > H^* = \frac{\phi_0}{\pi(2d^3l)^{1/2}}. \quad (7)$$

The field  $H^*$  can also be thought of as the crossover field.<sup>1)</sup> It is easy to verify that the expression for the crossover field of the sandwich differs only insignificantly from Eq. (4) for  $H_{cr}$  in superlattices, in particular for  $s \ll d$ :  $H_{cr} = \phi_0 / \pi(D^2ld)^{1/2} \rightarrow \phi_0 / \pi(d^3l)^{1/2}$  as  $s \rightarrow 0$ .

Just as for multilayer superlattices, we can define a characteristic “extrapolated” temperature  $\tilde{T}_c$  for a symmetric sandwich:<sup>8,12</sup>

$$\tilde{T}_c = T_c(1 - \xi^2/dl). \quad (8)$$

This expression is close to Eq. (3) for multilayer samples.

Thus, the mechanism that leads to crossover is qualitatively the same for superlattices and symmetric sandwiches. It is connected with changing the ratio between the size of the superconducting seed and the thickness of a single metallic layer of the layered structure. The values of the characteristic temperature  $\tilde{T}_c$  that the low-temperature square-root dependence of  $H_{c\parallel}(T)$  extrapolates to, and the crossover field, differ only minimally for the two systems under discussion—i.e., by only a factor of two. This difference is explained very simply: in superlattices, a superconducting seed in the range of  $2D$  behavior “adjoins” two metallic layers that are in the normal state through the thin interlayer, whereas in a sandwich it adjoins only one. Thus, the two systems agree with regard to the crossover mechanism and differ slightly in their mathematical expressions.

It should also be noted that for a superlattice<sup>8</sup> and a symmetric sandwich<sup>12</sup> a smooth and gradual transition from one dependence of  $H_{c\parallel}(T)$  to another is typical of the neighborhood of the crossover field.

The symmetric sandwich with thin metallic layers can be viewed as the limiting case of a superlattice with finite thickness. However, we observe that it exhibits a  $2D' - 2D$  instead of a  $2D' - 3D$  crossover; the former corresponds to a transition between two different two-dimensional states, and there is no region of  $3D$  behavior.

On the other hand, it is obvious that the crossover temperature for  $2D' - 3D$  in superlattices with a finite number of layers  $N$  is closer to  $T_c$ , the larger the total thickness  $L = ND$ . When this temperature lies in the range where the transition is resistive and washed out at  $H = 0$ , such crossovers are not seen and three-dimensional behavior is observed near  $T_c$ . For this reason, crossover is not observed for a symmetric sandwich made from two relatively thick layers.

A significantly different situation occurs for asymmetric sandwiches (see Figs. 3 and 4). For these sandwiches, the primary reason for crossover is the fact that the superconducting seed discontinuously jumps from one layer to another,<sup>12</sup> choosing the position corresponding to an absolute extremum of  $T_c(H)$ . Therefore, this type of crossover is characterized by a kink when we pass from one dependence to the other. An analogous crossover mechanism is characteristic for superlattices of  $S/S'$  type, where the same “hopping” of the superconducting seed from the  $S$ - to the  $S'$ -layer leads to an abrupt change in the temperature dependence of  $H_{c\parallel}(T)$  (the so-called Takahashi–Tachiki effect<sup>18</sup>). For this system, a sharp kink is actually observed at the crossover point,<sup>4,21,22</sup> along with a discontinuous change in the width of the resistive transition.<sup>22</sup> It is noteworthy that in several cases where the  $3D - 2D$  crossover temperature is close to  $T_c$  for the superlattices under consideration and the singularity of the temperature dependence of  $H_{c\parallel}$  associated with the Takahashi–Tachiki effect is most clearly evident, the dependences  $H_{c\parallel}(T)$  are essentially identical for superlattices and

for asymmetric sandwiches (compare, e.g., the data for Nb/NbZr superlattices in Fig. 1 of Ref. 21 with Fig. 4 of the present work).

Thus, dimensional crossover is a property not only of periodic monolayer systems but also of the “elementary bricks” from which the superlattice is built. Thus, in type  $S/I$  superlattices the elementary bricks are two identical superlattice films separated by the insulating layer. The properties of type  $S/S'$  superlattices can be modeled as assemblies of thin-layer asymmetric sandwiches.<sup>2)</sup>

From a comparison of the data of Fig. 1 for superlattices and Fig. 3 for sandwiches, it follows that the experimental observation of  $3D - 2D$  crossover cannot serve as evidence that the superlattice structure is ideal. In Ref. 8, it was shown that the phenomenon of dimensional crossover is stable against weak disruption of the periodicity. Comparison of the results for superlattices and for sandwiches indicates that large-scale disruption of the superstructure does not lead to elimination of crossover either. Thus, the observation of crossover based on the function  $H_{c\parallel}(T)$  can only indicate the presence of a layered structure, but not specific characteristics of the nonuniformity in the direction transverse to the layers.

However, there is a measurement method that allows us to unambiguously distinguish superlattices from sandwiches, and also ideal superlattices from defective ones. As we noted in the Introduction, it has been shown theoretically<sup>16</sup> and experimentally<sup>15</sup> that the function  $H_{c2}(\theta)$  for a superlattice, and especially the singularity in the value of the derivative of the critical field with respect to angle  $\beta = dH_{c2}/d\theta|_{\theta=0^\circ}$  near parallel orientation of the magnetic field relative to the layers, are very sensitive to irregularities in the superlattice. The derivative of the critical field with respect to angle is far more sensitive to restructuring of the spatial distribution of the order parameter than is the critical field itself. This clearly follows by analogy with the results of Saint-James<sup>24</sup> and Thompson<sup>25</sup> for single films, for which passage into the one-dimensional mixed state is accompanied only by a kink in the function  $H_{c\parallel}(T)$ , whereas the quantity  $\beta$  exhibits a distinct extremum.

Let us first compare the results for superlattices and sandwiches in the strong-field regime  $H > H_{cr}$ . According to Ref. 16, the function  $\beta(H)$  takes the following form for type  $S/I$  superlattices in large magnetic fields

$$\beta_{SL}(H) = -\frac{3\phi_0}{\pi d^2} \left\{ 1 - \frac{1}{4} \left( 6\frac{D^2}{d^2} + 1 \right) \left( \frac{H_{cr}}{H} \right)^4 \right\}, \quad (9)$$

$$H > H_{cr}.$$

This dependence, along with the experimental data for V/Si superlattices, is shown in Fig. 6. The agreement between experiment and Eq. (9) in the region under discussion is entirely satisfactory. In this range of magnetic fields, the influence of defects in the superstructure on the quantity  $\beta_{SL}$  is not appreciable, since the superconducting seed is localized in a single layer. The sole difference between ideal and defective superlattices is the fact that in the former, any position of the superconducting seed (in the absence of surface superconductivity) corresponds to the same free energy,

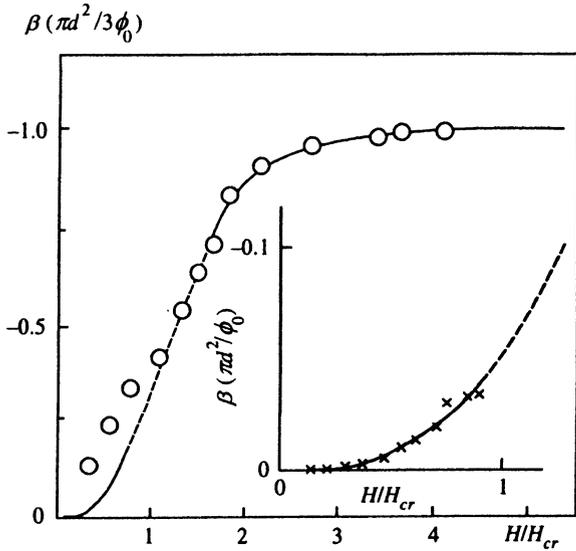


FIG. 6. Dependence of the normalized derivative of the critical field with respect to angle for  $\theta=0^\circ$  on the reduced field  $H/H_{cr}$  for a V/Si superlattice ( $D=330 \text{ \AA}$ ). The solid curves give the theoretical dependence corresponding to Eqs. (9), (11), the dashed curve interpolates between these expressions in the neighborhood  $H \sim H_{cr}$ . In the inset we give the initial portion of the dependence of  $\beta$  on  $H/H_{cr}$  for a Nb/Si superlattice ( $d=30 \text{ \AA}$ ,  $s=50 \text{ \AA}$ ). The solid curve is computed using Eq. (11).

whereas in the latter, it is advantageous for the seed to be localized in that layer for which the value of  $H_{c\parallel}$  is greatest, i.e., in the layer with the least value of  $d\xi$  (see Eq. (2) for the region of  $2D$  behavior). In this case, for superlattices that are close to ideal, the values of the quantity  $d$  obtained by measuring the angular dependence based on Eq. (9) and by measuring  $H_{c\parallel}(T)$  (Eq. (2)) are in very good agreement, whereas in defective superlattices they can disagree. A noticeable departure from the theory of Ref. 16 in the range of fields  $H > H_{cr}$  appears only for superlattices with very thin metallic layers. In Fig. 7 we present the results for a Mo/Si superlattice ( $d=30 \text{ \AA}$ ,  $s=25 \text{ \AA}$ ). It follows from this figure that in strong fields the function  $\beta_{SL}(H)$  tends toward saturation, but the value of  $\beta_{SL}$  at the plateau is very much smaller than its theoretical value. This discrepancy is not associated with the influence of defects in the superstructure, but rather with

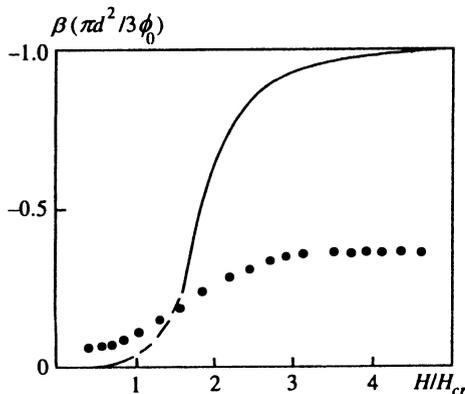


FIG. 7. Dependence of the normalized quantity  $\beta$  on  $H/H_{cr}$  for a Mo/Si superlattice ( $d=30 \text{ \AA}$ ,  $s=25 \text{ \AA}$ ).

the small value of  $d$ . Investigations of single ultrathin layers<sup>26</sup> have shown that for such small thicknesses of the superconducting layers the paramagnetic limit and spin-orbit interaction become important as well as the orbital effect, leading to an important change in the parallel critical field. These factors should also affect  $\beta$ .

For symmetric  $S/I/S$  sandwiches in the strong-field regime we obtain the dependence<sup>16</sup>

$$\beta_S(H) = -\frac{3\phi_0}{\pi d^2} \left[ 1 - \left( \frac{H_{cr}}{H} \right)^4 \right]^{1/2} \left[ 1 + 3 \left( \frac{H_{cr}}{H} \right)^4 \right]^{-1},$$

$$H > H_{cr}. \quad (10)$$

This dependence (and the theoretical dependence for the regime  $H < H_{cr}$ , which we address below) is shown in the inset to Fig. 5 by the solid curve. As in the case of superlattices, agreement between experiment and theory in this regime of fields is very good. From a comparison of Eqs. (9) and (10) it follows that the values of  $\beta$  at the plateau coincide for superlattices and for sandwiches for the same thicknesses of the metallic layers. However, this result is obvious and requires no detailed theoretical discussion, since in both cases we are in a purely two-dimensional situation, where the value of  $\beta$  for  $H \gg H_{cr}$  is determined by the localization of the superconducting seed to a single layer. The function  $\beta(H)$  reaches the plateau region smoothly both for superlattices and sandwiches. Thus, there are no significant qualitative differences for the two layered systems under discussion when  $H > H_{cr}$ .

An entirely different situation obtains in the weak magnetic field regime  $H < H_{cr}$ . Here the functions  $\beta(H)$  are completely different for superlattices and sandwiches. Furthermore, each of them is very sensitive to structural defects.

For an ideal superlattice, the value of  $\beta_{SL}$  in weak fields is exponentially small<sup>16</sup>:

$$\beta_{SL}(H) = -\frac{2^{7/4}\phi_0}{\pi^{1/4}D^2} \left( \frac{H}{H_{cr}} \right)^{1/4} \exp\left(-2\frac{H_{cr}}{H}\right), \quad H < H_{cr} \quad (11)$$

(see Fig. 6). It should be noted that in the weak-field regime the agreement between experiment and theory is often found to be much worse than when  $H > H_{cr}$ . There is nothing surprising in this, since the influence of defects in the superstructure is most important in this range of fields. Thus, for example, in the presence of even a single "defective" metallic layer, the function  $\beta_{SL}(H)$  in weak fields takes the form<sup>16</sup>

$$\beta_{SL}(H) = -\frac{\sqrt{2}\phi_0}{\pi^{5/4}D^2} \left( \frac{\Delta d}{d} \right)^{1/2} \left( \frac{H}{H_{cr}} \right)^{5/4}, \quad H < H_{cr} \quad (12)$$

and we have, in place of exponential behavior, close to a linear increase in  $\beta_{SL}(H)$ . In deriving Eq. (12), the defect under discussion was taken to be a superconducting layer with a thickness different by an amount  $\Delta d$  from the thickness  $d$  of all the other layers in the superlattice ( $\Delta d/d \ll 1$ ). The presence of other types of defects in the superstructure (e.g., a deviation in the thickness of one of the semiconductor interlayers from normal) should also lead to an increase in  $\beta_{SL}$  compared to its values for an ideal superlattice determined by Eq. (11). It is obvious that the deviation of the

experimental function  $\beta_{SL}(H)$  from (11) for the V/Si sample should be connected with just this imperfection of the superlattice. For a more regular structure (e.g., the Nb/Si superlattice shown in the inset to Fig. 6) good agreement with Eq. (11) is observed.

For a symmetric thin-film sandwich  $\beta_S(H)$  in weak fields is determined by the expression<sup>16</sup>

$$\beta_S(H) = -\frac{3\phi_0}{4\pi d^2} \left[ 1 - \left( \frac{H}{H_{cr}} \right)^2 \right]^{1/2}, \quad H < H_{cr}. \quad (13)$$

One difference in the behavior of  $\beta(H)$  for superlattices and sandwiches is that the value of  $\beta_S(0)$  in the limit  $H \rightarrow 0$  is finite for a sandwich, whereas for an infinite superlattice (both ideal and defective) as  $H$  decreases we have  $\beta_{SL} \rightarrow 0$ . A second significant difference between these layered systems is the fact that for superlattices  $\beta_{SL}$  increases monotonically with increasing  $H$  (Fig. 6), whereas for a symmetric sandwich this dependence is nonmonotonic, and at the crossover field  $\beta_S$  vanishes.

The experimental data for  $\beta_S(H)$  in fields  $H < H_{cr}$  for a symmetric sandwich (the inset to Fig. 5) agree only qualitatively with Eq. (13). As in the case of superlattices, the reason for this disagreement with theory is connected with the nonideality of the structures (in this case with the deviation from ideal symmetry). The influence of weak asymmetry on the behavior of  $\beta_S(H)$  is discussed in more detail in Ref. 13.

It follows from the experimental functions  $\beta(H)$  shown here for superlattices and sandwiches (Figs. 5 and 6) that, independent of the quality of the samples, a radical difference is observed in the behavior of the functions  $\beta(H)$  for these two layered systems in weak fields, in contrast to the behavior of the function  $H_{c||}(T)$ . Therefore, measurements of  $\beta(H)$  allow us to reliably determine the nature of the nonuniformities in layered systems.<sup>3)</sup>

For asymmetric sandwiches, when  $H > H_{cr}$  the quantity  $\beta_{S\infty} = 3\phi_0/\pi d_2^2$  is determined by a thickness  $d_2$  thinner than the cladding layers. In the strong-field regime, the behavior of this system naturally does not differ from that of symmetric sandwiches and superlattices. When  $H < H_{cr}$ , in contrast to the symmetric sandwiches, the quantity  $\beta_S$  is also constant and equals  $3\phi_0/\pi d_1^2$ .<sup>13</sup> At the crossover field a sharp discontinuity in  $\beta_S$  occurs.

In discussing  $2D' - 2D$  crossover for asymmetric thin-film sandwiches, we noted the similarity between this crossover and  $2D - 2D''$  crossover for  $S/S'$  type superlattices. For these two types of layered samples we observe certain similarities in the behavior of  $\beta(H)$  as well. According to the experimental data of Ref. 22, obtained from Nb/Nb<sub>0.5</sub>Zr<sub>0.5</sub> superlattices at the temperature for the Takahashi–Tachiki crossover, there is a discontinuity in  $\beta_{SL}$  that is in qualitative correspondence with the results of theoretical calculations for superlattices of this type.<sup>27</sup> A jump in  $\beta_S$  at the crossover point is observed in asymmetric sandwiches as well. From a comparison of these results it follows that in different systems with the same crossover mechanism (jumping of the superconducting seed from one layer to another) the singularity in  $\beta(H)$  at the field  $H_{cr}$  has the same character.

#### 4. CONCLUSION

In this paper we have presented our measurements of the temperature and angular dependence of critical magnetic fields for various layered superconducting systems. Among the samples we studied were superlattices of type S/I (Mo/Si, V/Si, Nb/Si), of type S/S' (V/Ta) and sandwiches of type S/I/S (V/Si/V).

We have discussed various types of dimensional crossover observed in multilayer periodic and nonperiodic systems:  $3D - 2D$  crossover in superlattices due to changes in the ratio between the size of a superconducting seed and the period of the superstructure;  $2D' - 3D$  crossover in finite superlattices connected with changes in the coherence length compared with the total thickness of the sample;  $2D' - 2D''$  crossover (the Takahashi–Tachiki effect), which is characteristic of type S/S' superlattices, associated with shifting of the seed from the S-layer to the S'-layer resulting in a transition between two different two-dimensional states. The same kind of crossover can be observed in superconducting sandwiches as well, where the character of the crossover is determined by the type of sandwich (symmetric or asymmetric) and the individual thicknesses of the cladding layers of the sandwich.

We have established a correlation between distinctive features of the functions  $H_{c||}(T)$  and  $\beta(H)$  for all types of periodic and nonperiodic layered structures. We have shown that in strong fields  $H \gg H_{cr}$  the behavior of  $\beta(H)$  is the same for superlattices and for sandwiches, whereas the character of this dependence in weak fields  $H < H_{cr}$  allows us to draw conclusions about the type of nonuniformity of a sample (symmetric or asymmetric) in the direction perpendicular to the layers and the presence of defects of its layered structure, without knowing anything about the overall structure of the sample.

Thus, the type of crossover that is characteristic of superlattices appears in the simplest layered system as well, consisting of two superconducting layers separated by another material. For this reason, a sandwich may be viewed as the limiting case of a superlattice with a finite number of layers. Differences of real importance between the different types of sandwiches and superlattices (with both infinite and finite lengths) are observed only in the weak-field behavior of the angular dependence of  $H_{c2}$  and  $\beta(H)$ .

Once we have investigated superconducting sandwiches, which from the point of view of simplicity of sample preparation are much more accessible and do not require high technology, we can predict rather precisely the properties of multilayer periodic systems consisting of the same materials, in particular: the parallel critical field in the region  $H > H_{cr}$ , the quantity  $\beta_{SL}$  at the plateau, and the crossover field and temperature. Additional details that justify this procedure can be found in our preceding article Ref. 13.

The authors are grateful to the International Science Foundation for partial support through grant No. U9M000.

<sup>1)</sup>In what follows, we will relabel the crossover field for the sandwich as  $H_{cr}$  for convenience, keeping in mind relation (7) if we are discussing sandwiches.

<sup>2)</sup>According to available experimental data for two-layer S/N and S/S' systems, the proximity effect can change only the critical parameters of the

superconducting layer (or the layer with the higher  $T_c$ ; see Ref. 23). It is obvious that when a suitable choice of the parameters of  $S$  and  $S'$  is made for the layers of  $S/S'$  and  $S/S'/S$  sandwiches, we should observe crossover-type effects at sufficiently low temperatures. Such experiments have not yet been carried out.

<sup>3</sup>Here the following remarks are necessary. For superlattices with a finite number of layers  $N$  in a very weak magnetic field ( $H \rightarrow 0$ ) the function  $\beta_{SL}(H)$  differs in form from that of an infinite superlattice, becoming nonmonotonic. Still another characteristic field appears—the field at which a vortex is introduced  $H_V$ . At this field value,  $\beta_{SL}$  reduces to zero, whereas  $\beta_{SL}(0)$  becomes finite.<sup>16</sup> An additional extremum is possible for sandwiches with the corresponding choice of parameters, whose origin is the same. Thus, a certain qualitative similarity can be observed between the functions  $\beta(H)$  for finite superlattices and sandwiches. This serves as one more confirmation of the fact that a sandwich is the limiting case of a superlattice with a finite number of layers. However, for superlattices the field  $H_V = 1.6H_{cr}/N^2$  is far smaller than the crossover field, so that this feature of  $\beta(H)$  is located far from  $H_{cr}$  and shifts toward  $H=0$  with increasing  $N$  as  $N^2$ . As for  $\beta(0)$ , for a finite superlattice this quantity can be comparable to  $\beta(0)$  for a symmetric sandwich only for large values of the anisotropy parameter and absolutely small  $N$  (see Ref. 16):  $\beta_{SL}(0) \approx 4(M/m)^{1/2}\beta_S(0)/N^2$ .

<sup>1</sup>D. E. Prober, R. E. Schwall, and M. R. Beasley, Phys. Rev. B **21**, 2717 (1980).

<sup>2</sup>I. Banerjee and I. K. Schuller, J. Low Temp. Phys. **54**, 501 (1984).

<sup>3</sup>K. Kanoda, H. Mazaki, N. Hosoito, and T. Shinjo, Phys. Rev. B **35**, 6736 (1987).

<sup>4</sup>M. J. Karkut, V. Matijasevic, L. Antognazza *et al.*, Phys. Rev. Lett. **17**, 1751 (1988).

<sup>5</sup>P. R. Broussard and T. H. Geballe, Phys. Rev. B **35**, 1664 (1987).

<sup>6</sup>S. T. Ruggiero, T. W. Barbie, and M. R. Beasley, Phys. Rev. B **26**, 4894 (1982).

<sup>7</sup>D. Neerincck, K. Temst, C. Van Hasendonck *et al.*, Phys. Rev. B **43**, 8676 (1991).

<sup>8</sup>L. I. Glazman, I. M. Dmitrenko, V. L. Tovazhnyanskii, Zh. Éksp. Teor. Fiz. **92**, 1461 (1987) [Sov. Phys. JETP **65**, 821 (1987)].

<sup>9</sup>S. Takahashi and M. Tachiki, Phys. Rev. B **33**, 4620 (1986).

<sup>10</sup>V. M. Gvozdkov, Fiz. Nizk. Temp. **16**, 5 (1990) [Sov. J. Low Temp. Phys. **16**, 1 (1990)].

<sup>11</sup>G. Deutscher and O. Entin-Wohlman, Phys. Rev. B **17**, 1249 (1978).

<sup>12</sup>L. I. Glazman, I. M. Dmitrenko, A. E. Kolin'ko *et al.*, Fiz. Nizk. Temp. **14**, 580 (1988) [Sov. J. Low Temp. Phys. **14**, 318 (1988)].

<sup>13</sup>V. G. Cherkasova, N. Ya. Fogel', and A. S. Pokhila, Fiz. Nizk. Temp. **20**, 1245 (1994) [Low Temp. Phys. **20**, 975 (1994)].

<sup>14</sup>V. L. Tovazhnyanskii, V. G. Cherkasova, and N. Ya. Fogel', Zh. Éksp. Teor. Fiz. **93**, 1384 (1988) [Sov. Phys. JETP **66**, 787 (1988)].

<sup>15</sup>V. L. Tovazhnyanskii, A. N. Stetsenko, A. I. Fedorenko *et al.*, Fiz. Nizk. Temp. **15**, 828 (1989) [Sov. J. Low Temp. Phys. **15**, 459 (1989)].

<sup>16</sup>L. I. Glazman, Zh. Éksp. Teor. Fiz. **93**, 1373 (1987) [Sov. Phys. JETP **66**, 780 (1987)].

<sup>17</sup>I. M. Dmitrenko, N. Ya. Fogel', V. G. Cherkasova *et al.*, Fiz. Nizk. Temp. **19**, 747 (1993) [Low Temp. Phys. **19**, 533 (1993)].

<sup>18</sup>S. Takahashi and M. Tachiki, Phys. Rev. B **34**, 3162 (1986).

<sup>19</sup>D. Saint-James, T. Sarma, and W. Thomas, *Type-II Superconductors* (Russ. transl. Mir, Moscow, 1970).

<sup>20</sup>A. A. Abrikosov, Zh. Éksp. Teor. Fiz. **47**, 720 (1964) [Sov. Phys. JETP **20**, 480 (1964)].

<sup>21</sup>Y. Kuwasawa, T. Tosaka, A. Uchiyama *et al.*, Physica C **175**, 187 (1991).

<sup>22</sup>Y. Kuwasawa, Y. Hayano, T. Tosaka *et al.*, Physica C **175**, 187 (1991).

<sup>23</sup>G. Deutscher and P. deGennes, in *Superconductivity*, R. D. Parks (ed.), Marcel Dekker, New York (1969), p. 1005.

<sup>24</sup>D. Saint-James and P. deGennes, Phys. Lett. **7**, 306 (1963).

<sup>25</sup>R. S. Thompson, Zh. Éksp. Teor. Fiz. **69**, 2249 (1975) [Sov. Phys. JETP **42**, 1144 (1975)].

<sup>26</sup>P. M. Tedrow and R. Meservey, Phys. Rev. B **8**, 5098 (1973).

<sup>27</sup>K. Takana, J. Phys. Soc. Jpn. **58**, 668 (1989).

Translated by Frank J. Crowne