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Dimensional Crossover in the Nb/Al_2O_3 Josephson-Coupled Multilayers

Manabu IKEBE, Yoshihisa OBI[1], Hiroyuki FUJISHIRO and Hiroyasu FUJIMORI[1]

Department of Materials Science and Technology, Faculty of Engineering, Iwate University, Morioka 020
[1]*Institute for Materials Research, Tohoku University, Sendai 980*

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The dimensional crossover from anisotropic three-dimensional to two-dimensional behavior in the temperature dependence of the parallel critical field has been studied for Nb/Al_2O_3 superconductive multilayers as a function of the structural modulation wavelength λ. The thicknesses of the superconductive Nb sublayer (dNb) and the Al_2O_3 tunneling barrier sublayer ($d\,Al_2O_3$) were varied up to 100 Å. In accord with the calculation due to Deutscher and Entin-Wohlman, the crossover occurs in the vicinity of T^*, where the perpendicular coherence length $\xi_\perp(T)$ satisfies the relation, $1.4\xi_\perp(T^*)=\lambda$, irrespective of the ratio λ/dNb.

[superconductivity, Nb/Al_2O_3 multilayer, superlattice, upper critical field, dimensional crossover]

§1. Introduction

The superconductors with layered structure show typical anisotropic and/or two-dimensional (2D) properties. Among those characteristic properties, the dimensional crossover in the parallel critical field ($H_{c2//}$) is the most conspicuous and has been the subject of several investigations.[1–5] This phenomenon is qualitatively understood in the following way. Near T_c, where the interlayer coherence length $\xi_\perp(T)$ is longer than the structural modulation wavelength λ, $H_{c2//}(T)$ is proportional to (T_c-T) in accord with the three-dimensional (3D) Ginzburg-Landau (GL) theory.[6] Below a certain temperature T^*, at which $\xi_\perp(T)$ becomes nearly equal to λ, $H_{c2//}$ behavior changes to that of a thin film and $H_{c2//}(T)$ diverges if we neglect the layer thickness and Pauli paramagnetic effect.[7] Such behavior of H_{c2} is designated as quasi-2D superconductivity. The dimensional crossover was initially observed in naturally occurring compounds, *e.g.*, intercalated transition metal dichalcogenides.[8] With the recent development of the deposition technique, the crossover has also been observed for many types of artificial multilayers, *e.g.*, Nb/Ge,[1] Nb/Cu,[2] Nb/Ti,[9] V/Ag[10] etc.

The artificial multilayer superconductors may be divided into two main types according to the nature of the interlayer coupling. One type is the superconductor/metal multilayer in which the proximity effect is responsible for the coupling.[11–14] The other type is the superconductor/insulator multilayer in which Josephson tunneling provides the coupling,[15,16] Experimentally, the dimensional crossover in the proximity-effect-coupled multilayers has been rather thoroughly investigated and detailed theoretical treatment has been given by Takahashi and Tachiki.[14,17,18] While the theoretical study on the layered superconductor was originated by Lawrence and Doniach[15] for the Josephson-coupled model, the experimental study on Josephson-coupled multilayers is scarce except for the pioneering work due to Ruggiero *et al.*[1] on Nb/Ge. In this paper, we examine the dimensional crossover of the Nb/Al_2O_3 multilayers. Because Al_2O_3 is a genuine insulator and Ge is a semiconductor, Nb/Al_2O_3 is expected to be a more ideal Josephson-coupled system than Nb/Ge. The synthesis and characterization of the Nb/Al_2O_3 multilayers have been described previously.[19,20] The critical field measurement has

been made resistively by use of a standard four-lead method.

§2. Theoretical Background

Layered superconductors can be considered as bulk or three-dimensional (3D) as long as the interlayer coherence length $\xi_\perp(T)$ is larger than the structural modulation wavelength λ by far. In this case, the upper critical fields parallel and perpendicular to the layer are given on the basis of the anisotropic GL theory by

$$H_{c2/\!/}(T)=\frac{\phi_0}{2\pi\xi_{/\!/}(T)\xi_\perp(T)} \tag{1}$$

$$H_{c2\perp}(T)=\frac{\phi_0}{2\pi\xi_{/\!/}(T)^2} \tag{2}$$

where $\xi_{/\!/}(T)$ is the intralayer coherence length and ϕ_0 is the flux quantum. In the anisotropic GL theory, effective GL masses parallel and perpendicular to the layer are defined by the following relation,

$$(m_\perp/m_{/\!/})^{1/2}=\left.\frac{dH_{c2/\!/}/dT}{dH_{c2\perp}/dT}\right|_{T\simeq T_c}, \tag{3}$$

In the Josephson-coupled model due to Lawrence and Doniach (LD),[15] layered superconductors are pictured as a stacked array of superconducting layers (with negligible thickness) weakly coupled through Josephson tunneling between adjacent layers. For field orientations except nearly parallel to the layer, the $H_{c2}(T)$ curves of LD theory are essentially the same as those predicted by the anisotropic GL theory. For $H_{c2/\!/}(T)$, however, the LD theory predicted quite anomalous behavior,

$$H_{c2/\!/}(T)\propto 1/[1-2\lambda^2/\xi_\perp^2(T)]^{1/2}. \tag{4}$$

Thus $H_{c2/\!/}$ diverges at a temperature T^* given by a relation

$$\xi_\perp(T^*)=\lambda/\sqrt{2}. \tag{5}$$

Klemm, Luther and Beasley[16] (KLB) extended the LD model and developed a microscopic theory. According to KLB, a characteristic parameter γ is defined by the following equation

$$\gamma=J^2\tau/T_c=\frac{4}{\pi}\left|\frac{\xi_\perp(0)}{\lambda/2}\right|^2, \tag{6}$$

where J is the tunneling energy and τ is the total electron scattering time. The parameter γ characterizes the degree of superconductive two-dimensionality, the smaller γ values indicating the stronger 2D characteristics. For $\gamma<1.76$, the KLB theory reproduced the $H_{c2/\!/}$ divergence and the Pauli paramagnetic limiting was taken account of to remove the unphysical divergence. In case that the interlayer Josephson-coupling is not so weak, the LD-KLB theory also reduces to the anisotropic GL theory. Then, in terms of the KLB theory, the mass ratio in eq. (3) is given by

$$(m_\perp/m_{/\!/})=2D/J^2\tau\lambda^2, \tag{7}$$

where D is the electron diffusion constant.

The KLB theory, however, neglected the superconducting layer thickness d_s. If we take into account the effect of finite d_s, the $H_{c2/\!/}$ divergence of the Josephson-coupled array should be limited by the $H_{/\!/}$ value of a single layer which is given by

$$H_{/\!/}=\frac{\sqrt{12}\,\phi_0}{2\pi\xi_{/\!/}d_s}. \tag{8}$$

Based on the KLB theory, Deutscher and Entin-Wohlman (DE)[21] calculated $H_{c2/\!/}(T)$ of Josephson-coupled layer superconductors keeping d_s finite and found that the crossover manifests itself as an upturn in the $H_{c2/\!/}(T)$ curve. DE pointed out that the condition for the dimensional crossover is given by

$$\lambda/\xi(T^*)\simeq 1.4 \tag{9}$$

independent of the λ/d_s value. By comparing eq. (9) with eq. (5) we notice that eq. (8) retains its validity to the limit of $d_s=0$, the limit treated by the LD-KLB theory. Thus we can also expect that the γ parameter defined by eq. (6) may be meaningful also in the finite d_s situation to characterize the superconductive dimensionality. In the following section, the prediction of eq. (9) due to Deutscher and Entin-Wohlman is examined for the Nb/Al_2O_3 artificial multilayers.

§3. Results and Discussion

The upper critical fields of Nb/Al_2O_3 multilayers with Nb and Al_2O_3 layer thicknesses (dNb and $d\,Al_2O_3$) ranging from 5 Å to 100 Å have been measured under the magnetic field

up to 90 kOe. For the present Nb/Al_2O_3 superlattice, dNb corresponds to d_s in the previous section and λ is given by the sum of dNb and $d Al_2O_3$,

$$\lambda = d\mathrm{Nb} + d\,Al_2O_3. \tag{10}$$

All the Nb/Al_2O_3 specimens with $d Al_2O_3 = 5$ Å showed anisotropic 3D superconductivity. All the specimens with $d Al_2O_3 \geqq 30$ Å showed 2D superconductivity. The effective mass ratio $m_\perp/m_{/\!/}$ of 2D multilayers is infinite and $H_{c2/\!/}(T)$ is described by eq. (8). Quasi-2D superconductivity is observable for $d Al_2O_3 = 10$ Å and 20 Å. In Figs. 1(a)~1(c), $H_{c2/\!/}(T)$ and $\xi_\perp(T)$ are plotted as a function of temperature T for typical specimens which show the crossover. The perpendicular coherence length $\xi_\perp(T)$ was estimated from the H_{c2} data by the use of the following relation based on the GL approximation:

$$\xi_\perp(T) = \phi_0^{1/2}\left[2\pi\frac{\mathrm{d}H_{c2\perp}}{\mathrm{d}T}(T_c - T)\right]^{-1/2} \times (m_{/\!/}/m_\perp)^{1/2}. \tag{11}$$

In the figures a horizontal line is drawn at $\xi_\perp = \lambda/1.4$ for each multilayer. According to DE, the cross point of the horizontal line and the $\xi_\perp(T)$ curve gives the crossover temperature T^* and $H_{c2/\!/}(T)$ should show an upturn near T^*. In Figs. 1(a)~1(c), we see that the upturn actually occurs slightly above the cross point. The prediction of Deutscher and Entin-Wohlman seems to be consistent with our experimental results.

The Nb/Al_2O_3 multilayers with $d Al_2O_3 =$ 10 Å show 3D superconductivity for dNb $\leqq$ 50 Å and quasi-2D superconductivity for dNb $\geqq$ 60 Å. An example of $H_{c2/\!/}(T)$ and $\xi_\perp(T)$ of the 3D specimen with $d Al_2O_3 = 10$ Å is shown in Fig. 2. The $\xi_\perp(T)$ curve of the 3D multilayers does not cross the horizontal $\lambda/1.4$ line, which is also consistent with the DE prediction. The Nb/Al_2O_3 multilayers with $d Al_2O_3 = 20$ Å show 3D superconductivity for dNb = 20 Å, quasi-2D superconductivity for 30 Å $\leqq d$Nb $\leqq$ 40 Å and 2D superconductivity for dNb $\geqq$ 60 Å. For an example of the 2D multilayer with $d Al_2O_3 = 20$ Å $H_{c2}(T)$ of Nb(60 Å)/Al_2O_3(20 Å) is shown in Fig. 3. These results indicate that the effect of the

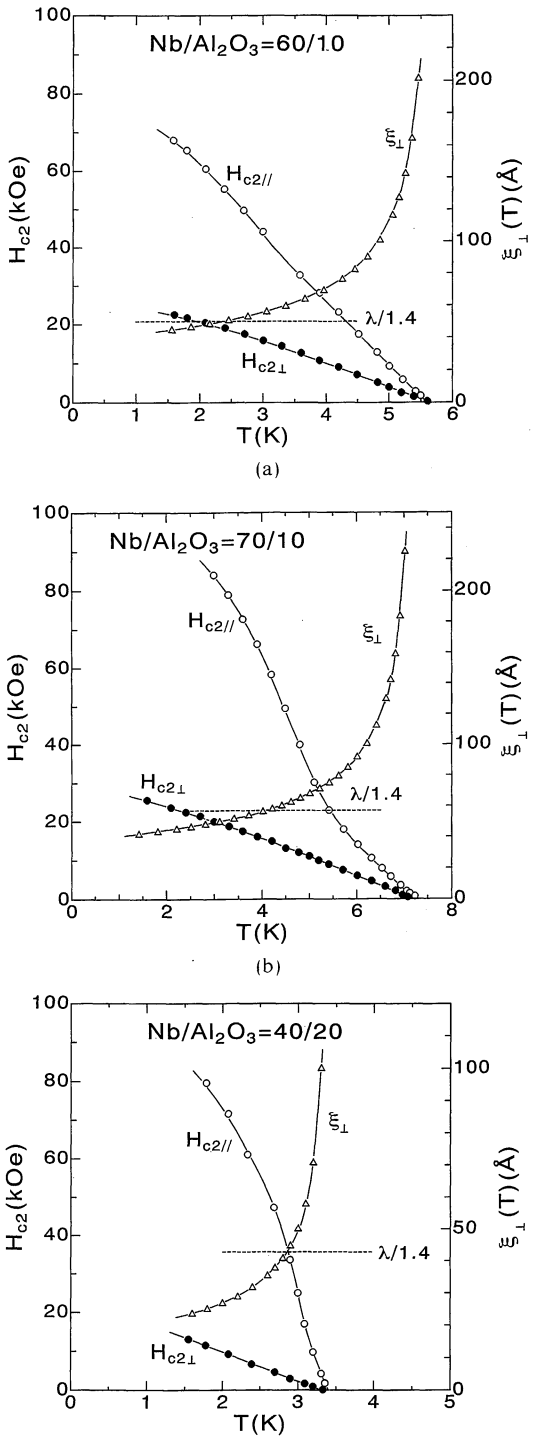

Fig. 1. $H_{c2/\!/}$ and $\xi_\perp(T)$ vs T for quasi-2D superconducting multilayers. (a) dNb/$d Al_2O_3$ = 60 Å/10 Å, (b) 70 Å/10 Å, (c) 40 Å/20 Å. $\xi(T)$ curves cross the horizontal dotted line drawn at $\lambda/1.4$ for each multilayer.

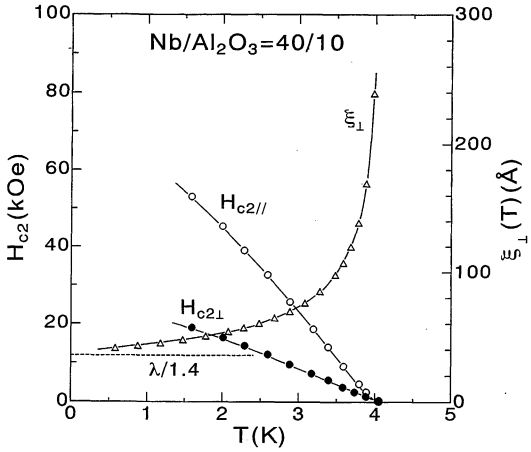

Fig. 2. An example of $H_{c2//}(T)$ and $\xi_\perp(T)$ for 3D multilayers, 40 Å/10 Å. $\xi(T)$ curves do not cross horizontal $\lambda/1.4$ line.

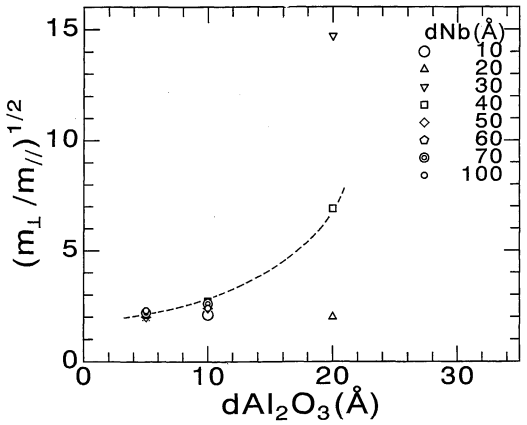

Fig. 4. The root of the mass ratio $(m_\perp/m_{//})^{1/2}$ vs $d\,Al_2O_3$. The drastic enhancement of $m_\perp/m_{//}$ occurs between $d\,Al_2O_3=10$ Å and 20 Å. The guide line for eyes is drawn for $d\,Nb=40$ Å.

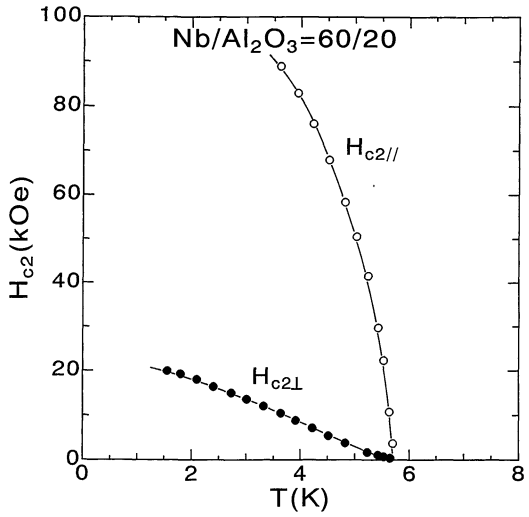

Fig. 3. An example of $H_{c2//}(T)$ and $H_{c2\perp}(T)$ for 2D multilayers, 60 Å/20 Å.

Al_2O_3 layer thickness on the dlmensionality of superconductivity is of essential importance as expected. But we must not overlook the fact that the effect of $d\,Nb$ is also very significant. From the present results, it is evident that the larger $d\,Nb$ values contribute to reduce the superconductive dimensionality. The origin which causes the $d\,Nb$ dependence of the dimensionality may be qualitatively understood in the following way. Since the number of Cooper pairs in a single Nb layer is proportional to $d\,Nb$, the tunneling of a larger number of Cooper pairs to the adjacent layer is necessary to result in sizable interlayer coupling between thicker Nb layers. Thus the Nb/Al_2O_3 multilayers with larger $d\,Nb$ tend to show lower superconductive dimensionality for a fixed $d\,Al_2O_3$, i.e., for a fixed tunneling energy J.

In Fig. 4 we plot the square root of the mass ratio $(m_\perp/m_{//})^{1/2}$ vs. $d\,Al_2O_3$ for 3D and quasi-2D multilayers. The ratio was determined from the temperature gradient of critical fields by the relation of eq. (3). As can be seen, the mass ratio sharply rises between $d\,Al_2O_3=10$ Å and 20 Å except for Nb(20 Å)/Al_2O_3(20 Å) which is a 3D superconductor. It should also be noticed that for $d\,Al_2O_3=20$ Å the superconductivity of multilayers with $d\,Nb \geqq 60$ Å is 2D with $(m_\perp/m_{//})^{1/2}$ values of infinity as is shown in Fig. 3. In the Nb/Ge system, the corresponding sharp of the ratio occurred in the vicinity of the Ge layer thickness $d\,Ge \simeq 30$ Å. In the Josephson-coupling model, $m_\perp/m_{//}$ is inversely proportional to J^2 as is given by eq. (6). For a fixed barrier layer thickness, the tunneling energy J is smaller in the Nb/Al_2O_3 system than the Nb/Ge system. Al_2O_3 is an intrinsic insulator, while Ge is a semiconductor. The larger gap in the electron energy band of Al_2O_3 may contribute to make the Al_2O_3 sublayer the more efficient tunneling barrier than the Ge sublayer.

We calculated the parameter γ defined by

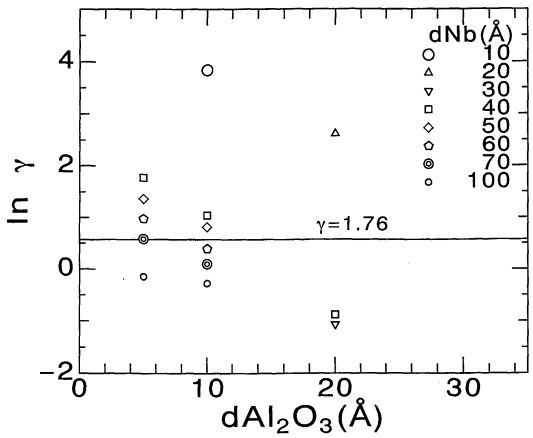

Fig. 5. $\log \gamma$ vs $d\mathrm{Al_2O_3}$. The horizontal line shows $\gamma=1.76$, by which superconductive dimensionality is divided into 3D (above the line) and quasi-2D (below the line) according to the KLB theory. This criterion is satisfied by all multilayers except Nb(100 Å)/$\mathrm{Al_2O_3}$(5 Å).

KLB using eq. (5) and $\log \gamma$ vs. $d\mathrm{Al_2O_3}$ is plotted for $d\mathrm{Al_2O_3}=5$, 10 and 20 Å in Fig. 5. We notice in the figure that all the sample with $\gamma>1.76$ show 3D superconductivity and all the sample with $\gamma<1.76$ show quasi-2D superconductivity in accord with the KLB theory. Only exception is Nb(100 Å)/$\mathrm{Al_2O_3}$(5 Å) with 3D superconductivity, the γ value of which is 0.95. From measurements of general transport properties, we have earlier suggested that $d\mathrm{Al_2O_3}=5$ Å may be too small to form well-defined tunneling barriers in the Nb/$\mathrm{Al_2O_3}$ system, though the existence of a layer-like modulated structure was verified by the satellite peaks of the small angle X-ray diffraction.[19] The discrepancy of the KLB γ value criterion for Nb(100 Å)/$\mathrm{Al_2O_3}$(5 Å) may be attributable to the imperfect barrier formation.

Finally we summarize in Table I several superconducting material parameters of the Nb/$\mathrm{Al_2O_3}$ multilayers discussed in this paper.

§4. Conclusion

1. The dimensional crossover has been studied for the Josephson coupled Nb/$\mathrm{Al_2O_3}$ multilayers by varying the ratio of the superconducting Nb layer thickness dNb to the structural modulation wavelength λ. The crossover occurs, independent of λ/dNb, in the vicinity of T^* where the perpendicular coherence length $\xi_\perp$ is equal to $\lambda/1.4$. This result is in agreement with the calculation due to Deutscher and Entin-Wohlman.
2. We have experimentally confirmed that the crossover temperature T^* depends upon not only the tunneling barrier thickness ($d\mathrm{Al_2O_3}$) but also the superconducting layer thickness (dNb).
3. The effective mass ratio $m_\perp/m_{/\!/}$ of the Nb/$\mathrm{Al_2O_3}$ multilayer sharply increases between $d\mathrm{Al_2O_3}=10$ Å and 20 Å. In the Nb/Ge

Table I. Superconducting parameters of Nb/$\mathrm{Al_2O_3}$ multilayers.

dNb/$d\mathrm{Al_2O_3}$ (Å/Å)	T_c (K)	$dH_{c2\perp}/dT$ (kOe/K)	$dH_{c2/\!/}/dT$ (kOe/K)	$(m_\perp/m_{/\!/})^{1/2}$	$\xi_\perp(0)$ (Å)	γ	$\ln\gamma$	$\xi_{/\!/}(0)$ (Å)	Dimension
40/5	3.496	9.9	20.0	2.0	48.2	5.85	1.17	97.5	3D
50/5	4.395	8.0	16.1	2.0	48.0	3.88	1.36	96.7	3D
60/5	4.825	7.4	15.2	2.1	46.7	2.63	0.97	96.0	3D
70/5	5.539	6.2	13.7	2.2	44.3	1.78	0.58	97.9	3D
100/5	6.599	5.2	11.8	2.3	43.1	0.86	−0.15	97.9	3D
10/10	1.810	11.8	24.2	2.1	60.5	46.6	3.84	124.1	3D
40/10	4.080	8.0	21.5	2.7	37.3	2.83	1.04	100.4	3D
50/10	5.000	7.2	17.3	2.4	39.8	2.24	0.81	95.6	3D
60/10	5.661	6.3	16.0	2.5	37.8	1.48	0.39	96.1	q-2D
70/10	7.217	4.8	12.5	2.6	37.4	1.11	0.10	97.5	q-2D
100/10	6.979	4.6	11.0	2.4	42.3	0.75	−0.28	101.2	q-2D
20/20	1.873	9.7	19.8	2.0	65.9	13.8	2.62	134.6	3D
30/20	3.013	3.0	44.0	14.7	13.0	0.34	−1.07	120.0	q-2D
40/20	3.398	7.0	48.0	6.9	17.1	0.41	−0.88	120.0	q-2D

system due to Ruggiero *et al.*,[1] the corresponding increase of $m_\perp/m_{/\!/}$ was observed at about $d\,\mathrm{Ge} \simeq 30$ Å. This results indicate that the Al_2O_3 insulator layer forms more efficient tunneling barrier in Nb based superlattice than the Ge semiconductor layer.

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References

1) S. T. Ruggiero, T. W. Barbee, Jr. and M. R. Beasley: Phys. Rev. B**26** (1982) 4894.
2) C. S. L. Chun, G. G. Zheng, J. L. Vincent and I. K. Schuller: Phys. Rev. B**29** (1984) 4915.
3) I. Banerjee and I. K. Schuller: J. Low Temp. Phys. **54** (1984) 501.
4) J. L. Cohn, J. J. Lin, F. J. Lamelas, H. He, R. Clarke and C. Uher: Phys. Rev. B**38** (1988) 2326.
5) D. Neerinck, K. Temst, C. V. Haesendonck, Y. Bruynseraede, A. Gilabert and I. K. Schuller: Phys. Rev. B**43** (1991) 8676.
6) V. L. Ginzburg and L. D. Landau: Zh. Eksp. Teor. Fiz. **20** (1950) 1064.
7) K. Maki: Phys. Rev. **148** (1966) 362.
8) See, for example, D. E. Proker, R. E. Schwall and M. R. Beasley: Phys. Rev. B**21** (1980) 2717; M. Ikebe and Y. Muto: Synthetic Metals **5** (1983) 229.
9) Y. Obi, M. Ikebe, Y. Fukumoto, Y. Muto and H. Fujimori: *JJAP Ser.1 Superconducting Materials*, ed. S. Nakajima and H. Fukuyama (Jpn. J. Appl. Phys., Tokyo, 1988) p. 129.
10) K. Kanoda, H. Mazaki, T. Yamada, N. Hosoito and T. Shinjo: Phys. Rev. B**33** (1986) 2052.
11) P. G. de Gennes and E. Guyon: Phys. Lett. **3** (1963) 168.
12) N. R. Werthamer: Phys. Rev. **132** (1963) 2440.
13) P. G. de Gennes: Rev. Mod. Phys. **36** (1964) 225.
14) S. Takahashi and M. Tachiki: Phys. Rev. B**33** (1986) 4620.
15) W. E. Lawrence and S. Doniach: *Proc. 12th Int. Conf. Low Temperature Physics, 1970*, ed. E. Kanda (Academic Press of Japan, Tokyo, 1971) p. 361.
16) R. A. Klemm, A. Luther and M. R. Beasley: Phys. Rev. B**12** (1975) 877.
17) M. Tachiki and S. Takahashi: Physica **135B** (1985) 178.
18) M. Tachiki and S. Takahashi: Physica C**153–155** (1988) 1702.
19) Y. Kamiguchi, M. Ikebe, Y. Obi and H. Fujimori: Jpn. J. Appl. Phys. **30** (1991) 945.
20) Y. Kamiguchi, Y. Obi, M. Ikebe and H. Fujimori: *Proc. the MRS Int. Meet. Advanced Materials, 1988*, ed. M. Doyama, S. Somiya and R. P. H. Chang (Materials Research Society, Pittsburgh, Pennsylvania, 1989) Vol. 10, p. 3.
21) G. Deutscher and O. Entin-Wohlman: Phys. Rev. B**17** (1978) 1249.