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Calculation of the upper critical field of V/Ag and Nb/Cu superlattices

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The Takahashi-Tachiki equations, describing the critical properties of proximity-effect systems in the dirty limit, are solved exactly using the full eigenfunction expansion. Both parallel and perpendicular critical fields are calculated. The theory is applied to experimental data of Kanoda *et al.* for V/Ag and of Chun *et al.* for Nb/Cu, using the T_c of the superconducting material and the diffusion coefficients of both materials as fit parameters. For the three-dimensional (3D) systems the fits compare nicely with the experimental results, but for 2D systems this is not always the case. It is found that the V and Nb critical temperatures necessary to fit the data can be larger than the corresponding bulk critical temperatures. This contrasts with what has been observed for single V and Nb films. Attempts to remedy this by choosing unconventional system parameters and using boundary conditions for less transparent interfaces turn out to be only limitedly successful. In the light of these anomalies earlier less complete calculations are reconsidered. It turns out that several assertions are superficial and cannot be affirmed by the results of the present work.

I. INTRODUCTION

In recent years the proximity effect in metallic multilayers has been subjected to detailed research, both experimentally and theoretically.¹ Advances in thin-film technology enabled the manufacturing of high-quality multilayer samples with layer thicknesses down to 15 Å. These were found to exhibit a variety of features that could be understood qualitatively by considering the layered nature of the samples.

A great deal of the experimental activities were focused on the Nb/Cu system. Banerjee *et al.*² measured the layer-thickness dependence of the multilayer critical temperature. In subsequent articles³⁻⁵ of Schuller and co-workers this work was extended to include measurements of the upper critical field H_{c2} . In these studies the layered nature of the samples had a clear manifestation in the dimensional crossover that was observed in the temperature dependence of the parallel upper critical field $H_{c2,\parallel}$. A system that is expected to be comparable to Nb/Cu is the V/Ag multilayer, whose two constituent metals belong to the same groups of the periodic system. Kanoda *et al.*^{6,7} studied the upper critical field properties of V/Ag experimentally. For intermediate layer thicknesses $H_{c2,\parallel}$ showed a dimensional crossover that looked qualitatively the same as that for Nb/Cu. The two-dimensionality of these multilayers was also reflected in $H_{c2,\perp}$ in terms of a positive second derivative of this quantity versus temperature. A few attempts were made to compare Nb/Cu results to the proximity effect theories available at the time. Banerjee *et al.*² analyzed their own T_c measurements using the Werthamer approximation⁸ and the Cooper-de Gennes limit.^{9,10} Biagi, Kogan, and Clem¹¹ tried to fit the outcome of the Werthamer theory to measurements of the perpendicular upper critical field. However, for the dimensional cross-over in $H_{c2,\parallel}$ no comparison with theory has been made as yet to our knowledge.

A great leap forward in the camp of the theorists was made in 1986.^{12,13} That year Takahashi and Tachiki¹² presented an advanced theory of the proximity effect in metallic multilayers. In fact, up to then the Werthamer approximation⁸ and the Cooper-de Gennes limit^{9,10} were the only descriptions available. But the Werthamer approximation, while working well for thick-layer systems, always overestimates T_c and H_{c2} when the layer thickness decreases.¹³ On the other hand, the Cooper-de Gennes limit gives the correct T_c in the thin-layer limit, but yields an underestimation for layers of finite thickness. The Takahashi-Tachiki formalism was shown¹³ to bridge the gap between these limiting behaviors. Exact solutions of the Takahashi-Tachiki equations were computed by Auvil and Ketterson¹⁴ for the case of zero magnetic field. The results were used to reconsider the analysis of Banerjee *et al.*² of multilayer T_c measurements.

Radović, Ledvij, and Dobrosavljević-Grujić¹⁵ developed a formalism that was shown¹³ to be equivalent to that of Takahashi and Tachiki in the limit that one of the constituent metals has a vanishing T_c , as is the case for Nb/Cu. They applied it to a model system for zero and perpendicular magnetic field only. Simultaneously, Takanaka¹⁶ performed calculations in which the Takahashi-Tachiki equations were solved exactly, for both parallel and perpendicular fields. Again, only model systems were investigated and no comparison was made to any real system.

Generally speaking, the effort to compare theory with measurements has been limited.¹ Many of the experimental data were presented without reference to theory, whereas many of the theoretical exertions restricted themselves to model systems. The comparative studies that have been made were incomplete in the sense that the three critical quantities T_c , $H_{c2,\perp}$, and $H_{c2,\parallel}$ were never treated together. Moreover, Takahashi and Tachiki¹² presented their results by comparing only reduced quan-

tities. Interestingly, in subjecting earlier studies to a supplementary analysis, striking inconsistencies show up, which could not be noticed at the time due to their incompleteness. It comes out that in order to check the status and applicability of the proximity-effect theory a full approach is indispensable. It is the aim of the present paper to meet this requirement.

This paper is organized as follows. After an introduction to the formalism of Takahashi and Tachiki in Sec. II, in Sec. III a critical review is given of previous studies. Subsequently, Secs. IV and V give a detailed comparison of the exact solution of the Takahashi-Tachiki equations with experimental data. We do not compare reduced quantities, for which satisfactory fits can more easily be obtained, but always consider the absolute values of T_c , $H_{c2,\perp}$, and $H_{c2,\parallel}$. The measurements that will be analyzed concern the V/Ag system⁷ and the Nb/Cu system.⁵ The main point of attention is the problem of the input parameters that one should use in order to bring the theory into agreement with experiment and how these parameters relate to their equivalents for a bulk metal.

II. THEORY

The starting point of the Takahashi-Tachiki theory^{12,13} is Gorkov's linearized integral equation for the pair potential¹⁷

$$\Delta(\mathbf{r}) = \int K(\mathbf{r}, \mathbf{r}') \Delta(\mathbf{r}') d^3 r' . \quad (1)$$

The kernel $K(\mathbf{r}, \mathbf{r}')$ can be expanded as

$$K(\mathbf{r}, \mathbf{r}') = V(\mathbf{r}) kT \sum_{\omega} Q_{\omega}(\mathbf{r}, \mathbf{r}') , \quad (2)$$

which contains a position-dependent BCS electron-electron interaction coupling constant $V(\mathbf{r})$ and a summation over discrete frequencies $\omega = (2n+1)\pi kT$. The summation is restricted to frequencies $|\omega| \leq \omega_D$, ω_D being the Debye frequency. According to Takahashi and Tachiki, $Q_{\omega}(\mathbf{r}, \mathbf{r}')$ can be derived from a set of coupled differential equations, including diamagnetic and paramagnetic effects. Auvil, Ketterson, and Song¹⁸ generalized these equations to include also magnetic impurity scattering and spin-orbit scattering. For nonmagnetic dirty type-II superconductors, however, these reduce to the single Green's-function-like differential equation

$$[2|\omega| + L(\nabla)] Q_{\omega}(\mathbf{r}, \mathbf{r}') = 2\pi N(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}') , \quad (3)$$

where $N(\mathbf{r})$ is the position-dependent density of states at the Fermi energy. The differential operator $L(\nabla)$ is given by

$$L(\nabla) \equiv -\hbar D(\mathbf{r}) \left[\nabla - \frac{2ie \mathbf{A}(\mathbf{r})}{\hbar c} \right]^2 , \quad (4)$$

$D(\mathbf{r})$ being the position-dependent electronic diffusion constant and $\mathbf{A}(\mathbf{r})$ the vector potential of the applied magnetic field. For multilayers, the three functions $N(\mathbf{r})$, $V(\mathbf{r})$, and $D(\mathbf{r})$ are assumed to be constant within a single material, making discontinuous jumps at the interfaces.

We now define the pair function $F(\mathbf{r}) \equiv \Delta(\mathbf{r})/V(\mathbf{r})$. It was the idea of Takahashi and Tachiki to solve Eqs. (1)

and (3) by using an expansion of $Q_{\omega}(\mathbf{r}, \mathbf{r}')$ and $F(\mathbf{r})$ in terms of the eigenfunctions $\psi_{\lambda}(\mathbf{r})$ of the operator $L(\nabla)$, with corresponding eigenvalues ε_{λ} :

$$F(\mathbf{r}) = \sum_{\lambda} c_{\lambda} \psi_{\lambda}(\mathbf{r})$$

and

$$Q_{\omega}(\mathbf{r}, \mathbf{r}') = \sum_{\lambda, \lambda'} a_{\lambda\lambda'}^{\omega} \psi_{\lambda}^*(\mathbf{r}') \psi_{\lambda}(\mathbf{r}) . \quad (5)$$

The eigenfunctions have to obey the de Gennes boundary conditions¹⁰ demanding the continuity of $\psi_{\lambda}(\mathbf{r})/N(\mathbf{r})$ and $D(\mathbf{r})[\nabla - 2ie \mathbf{A}(\mathbf{r})/\hbar c] \psi_{\lambda}(\mathbf{r})$. The orthogonality and closure properties read as

$$\int d^3 r \psi_{\lambda}^*(\mathbf{r}) \frac{1}{N(\mathbf{r})} \psi_{\lambda'}(\mathbf{r}) = \delta_{\lambda\lambda'} \quad (6)$$

and

$$\sum_{\lambda} \psi_{\lambda}^*(\mathbf{r}) \psi_{\lambda}(\mathbf{r}') = N(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}') , \quad (7)$$

reflecting the discontinuous nature of $N(\mathbf{r})$. Substitution of (5) in Eqs. (1) and (3) finally leads to a matrix equation for the coefficients c_{λ} :

$$c_{\lambda} = \sum_{\lambda'} \left[2\pi kT \sum_{\omega} \frac{1}{2|\omega| + \varepsilon_{\lambda}} \right] V_{\lambda\lambda'} c_{\lambda'} , \quad (8)$$

where $V_{\lambda\lambda'}$ is the matrix element $\langle \psi_{\lambda} | V | \psi_{\lambda'} \rangle$. When the secular equation

$$\det \left| \delta_{\lambda\lambda'} - 2\pi kT \sum_{\omega} \frac{1}{2|\omega| + \varepsilon_{\lambda}} V_{\lambda\lambda'} \right| = 0 \quad (9)$$

is satisfied, there are nontrivial solutions for the coefficients c_{λ} and consequently for the pair function $F(\mathbf{r})$. The highest temperature for which such a solution exists is the field-dependent critical temperature. The present authors showed¹³ that the frequency summation appearing in (9) can be evaluated exactly in terms of the digamma function $\Psi(x)$, using the identity

$$2\pi kT \sum_{\omega} \frac{1}{2|\omega| + \varepsilon_{\lambda}} = \Psi \left[\frac{\omega_D}{2\pi kT} + \frac{\varepsilon_{\lambda}}{4\pi kT} + 1 \right] - \Psi \left[\frac{\varepsilon_{\lambda}}{4\pi kT} + \frac{1}{2} \right] . \quad (10)$$

The de Gennes boundary conditions mentioned above do not account for the possible effect of a finite transparency of the metal-metal interfaces. In the present work, however, the consequences of this will be discussed as well. Kuprianov and Lukichev¹⁹ have shown how this effect of electron scattering at the interfaces can be included by modifying the boundary conditions. Rewritten to the above formulation,^{20,21} these were found to read as

$$D(\mathbf{r}) \frac{\partial}{\partial z} \psi_{\lambda}(\mathbf{r}) \Big|_{\mathbf{r}=\mathbf{r}^+} = D(\mathbf{r}) \frac{\partial}{\partial z} \psi_{\lambda}(\mathbf{r}) \Big|_{\mathbf{r}=\mathbf{r}^-} = \frac{1}{e^2 R_B} \left[\frac{\psi_{\lambda}(\mathbf{r}^+)}{N(\mathbf{r}^+)} - \frac{\psi_{\lambda}(\mathbf{r}^-)}{N(\mathbf{r}^-)} \right] , \quad (11)$$

where R_B is the effective resistance of the boundary, multiplied by its area. The symbols \mathbf{r}^- and \mathbf{r}^+ denote the vectors $(x, y, z - \varepsilon)$ and $(x, y, z + \varepsilon)$, respectively, where (x, y, z) is the vector that points at the interface, ε is an infinitesimal positive quantity, and z is assumed to be the direction perpendicular to the interfaces, that is, the direction in which $D(\mathbf{r})$ and $N(\mathbf{r})$ show discontinuities. The vector potential $\mathbf{A}(\mathbf{r})$ is assumed to have no z component. Equation (11) reduces to the de Gennes boundary conditions when the first and second members on the left-hand side are much smaller than either of the terms on the right-hand side. Since $\partial\psi_\lambda(\mathbf{r})/\partial z$ is of the order $\psi_\lambda(\mathbf{r})/\xi_i$, with ξ_i the smallest coherence length¹³ and where i can refer to either of the metals adjacent to the interface, this is true when $\xi_i \gg e^2 N_i D_i R_B$. From the microscopic transport equations it can be derived¹⁰ that R_B is of the order $\rho_i l_i / t_i$, where l_i is the electron mean free path and t_i is the transmission coefficient. Since $\rho_i = 1/e^2 N_i D_i$, this implies that the de Gennes boundary conditions are valid when $\xi_i \gg l_i / t_i$. When t_i is of the order of unity this condition is precisely the dirty-limit requirement, assumed to be satisfied in the present theory, but when t_i is much smaller than unity this condition may also be violated. Then Eq. (11) has to be used instead of the de Gennes boundary conditions. Note that R_B is finite even for a unit transmission coefficient, while the de Gennes boundary conditions are exact only for $R_B = 0$. Therefore the latter are an approximation even for the case of fully transparent boundaries and it might be advisable to use expression (11) for all cases.

III. CRITICAL REVIEW

This section is a review of previous publications in which proximity-effect theories are compared with experiment. These studies concern the Nb/Cu system only. For V/Ag no comparison is available as yet. The analysis is focused on the material parameters that have been employed by the various authors. As was seen in Sec. II, the theory requires four parameters to characterize a metal. These are the density of states at the Fermi energy, N , the BCS coupling constant V , the electronic diffusion coefficient D , and the Debye temperature Θ_D . The theory does not account for the two metals having different Debye temperatures. In all cases where this quantity is needed 295 K is used for both Nb and Cu. This is the average of the two individual Debye temperatures, which are $\Theta_{D, \text{Nb}} = 275$ K and $\Theta_{D, \text{Cu}} = 315$ K. Sometimes $T_{c, \text{Nb}}$ is used as a material parameter instead of V_{Nb} . In that case V_{Nb} is chosen such as to yield the given niobium T_c . This can be done since the theory defines a unique relationship between T_c , V , and N for a bulk material.

In 1982 Banerjee *et al.*² studied the dependence of the multilayer critical temperature T_c on the individual layer thicknesses. As expected theoretically, the proximity effect reduced T_c when the layer thicknesses were decreased. However, the amount of reduction was larger than could be explained by either the Werthamer theory⁸ or the Cooper-de Gennes limit.^{9,10} This led Banerjee *et al.* to the conclusion that the critical temperature of

the niobium itself is a decreasing function of its layer thickness. This argument was based on the assumption that the electronic mean free path is limited by the layer thickness. For increasingly thin layers this causes a suppression of the mean free path that smears out the electronic density of states. For niobium this leads to a reduction of the density of states at the Fermi energy and, consequently, to a lower T_c . This effect has been conjectured by Wolf, Kennedy, and Nisendorf²² for single niobium films.

To carry out the Werthamer calculations, Banerjee *et al.*² proposed a set of system parameters that was based on measurements of the normal-state resistivity ρ , the coefficient of normal-state electronic specific heat γ , and the electronic mean free path l . Using free-electron formulas their values for ρ , γ , and l can be shown to lead to the parameter values $N_{\text{Nb}} = 12.0 \times 10^{47} / \text{J m}^3$ and $N_{\text{Cu}} = 1.98 \times 10^{47} / \text{J m}^3$ and to the following recipes for determining the diffusion coefficients:

$$D_{\text{Nb}} = \min \left[1, \frac{d_{\text{Nb}}}{160 \text{ \AA}} \right] \times 3.47 \text{ cm}^2/\text{s}$$

and

$$D_{\text{Cu}} = \min \left[1, \frac{d_{\text{Cu}}}{2000 \text{ \AA}} \right] \times 65.7 \text{ cm}^2/\text{s},$$

where d_{Nb} and d_{Cu} denote the respective layer thicknesses. The minimal-value operators appearing in these equations reflect the fact that the mean free path is limited by the layer thickness when the latter is less than the bulk mean free path. The diffusion coefficient itself is proportional to the mean free path according to the free-electron formula $D = \frac{1}{3} v_F l$. As can be inferred from Eq. (12), bulk mean free paths of 160 Å for niobium and 2000 Å for copper were used by Banerjee *et al.* A vanishing T_c was assumed for copper, whereas $T_{c, \text{Nb}}$ was used as a free parameter to fit the multilayer critical temperature to the experimental data. This fit resulted in a $T_{c, \text{Nb}}$ that decreases for decreasing layer thickness. This decrease is somewhat less than in the plot of Wolf, Kennedy, and Nisendorf²² for the T_c of single niobium films, but it is in qualitative agreement with it. The bulk critical temperature for niobium was 8.91 K.

To determine the actual status of this seemingly satisfactory result it is necessary to draw attention to the limitations of the approach that has been followed. The first is that the Werthamer approximation is used. In the description of Banerjee *et al.* the observed decrease in the multilayer T_c is attributed to two mechanisms: the proximity effect on the one hand and on the other hand the decrease of the niobium T_c . The Werthamer theory underestimates the decrease due to the proximity effect and therefore necessarily overestimates the decrease due to the niobium T_c . In a more exact approach a lesser decrease of $T_{c, \text{Nb}}$ would be needed to fit the data. A second limitation is that the result depends strongly on the choice of the material parameters, in particular, the diffusion coefficients. In order to judge the reasonableness of these parameters it is an absolute necessity to in-

clude the theoretical predictions for the upper critical fields into the argument. Since this has not been done by the authors, it is premature to draw conclusions concerning the correspondence between theory and experiment. Another choice of parameters might be needed to fit upper critical field data, parameters, as we will show, that can lead to qualitatively different behavior of the niobium critical temperature.

In later work³⁻⁵ the anisotropic behavior of the upper critical field H_{c2} of Nb/Cu was mapped out extensively. It was found that for intermediate layer thicknesses the parallel upper critical field $H_{c2,\parallel}$ shows a clearly observable dimensional crossover from anisotropic three-dimensional (3D) behavior at higher temperatures to 2D thin-film-like behavior at lower temperatures. These were purely experimental articles. No comparison was made with theory. Figure 1 is a comparison of these experiments with the analysis of Banerjee *et al.*² It shows the data of Chun *et al.*⁵ and the exact solution of the Takahashi-Tachiki equations for the above set of parameters. The T_c of niobium was taken to be 8.4 K, consistent with the decrease of the niobium T_c that was inferred by Banerjee *et al.*² It is seen that these parameters do not reproduce the measurements in a satisfactory way.

A second comparison between theory and experiment was given by Biagi, Kogan, and Clem.¹¹ They analyzed the perpendicular upper critical field $H_{c2,\perp}$ of the Nb/Cu system in the framework of the Werthamer theory. It was found that the theory could be brought into agreement with the data of Banerjee and Schuller⁴ only when the mean free paths of both the niobium and the copper were assumed to be considerably smaller than the estimates obtained from longitudinal resistivity measurements. It was also shown that the experimentally observed positive curvature of $H_{c2,\perp}(T)$ can be reproduced using Werthamer theory.

To judge the significance of these findings we will subject their treatment of the system parameters to a supplementary analysis and reconsider their findings from the viewpoint of a more complete approach. The authors studied two Nb/Cu multilayer systems with equal layer

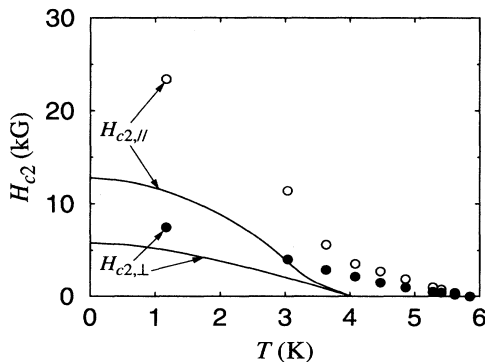


FIG. 1. Upper critical field curve of Nb(171 Å)/Cu(376 Å). The lines show the exact solution of the Takahashi-Tachiki equations, using the material parameters of Banerjee *et al.* (Ref. 2). The circles show the experimental data of Chun *et al.* (Ref. 5).

thicknesses $d = d_{\text{Nb}} = d_{\text{Cu}}$, d being either 420.5 or 171.5 Å. Copper was assumed to have zero T_c , whereas for $T_{c,\text{Nb}}$ layer-thickness-dependent values were used. The latter were taken from the fit to multilayer T_c data made by Banerjee *et al.*² The three remaining independent parameters, D_{Cu} , D_{Nb} , and the ratio $N_{\text{Cu}}/N_{\text{Nb}}$, were used to fit the theoretical results to the $H_{c2,\perp}$ data of Banerjee and Schuller.⁴ The results are shown in Table I. With these parameters the theoretical curves coincide with the $H_{c2,\perp}$ data over the whole temperature range.

The diffusion coefficients were analyzed in terms of the corresponding mean free paths. These were determined by making use of the free-electron formula $D = \frac{1}{3}v_F l$ and the Fermi velocities $v_{F,\text{Nb}} = 2.73 \times 10^7$ cm/s and $v_{F,\text{Cu}} = 1.57 \times 10^8$ cm/s. Table I shows that the resulting mean free paths are in all cases smaller than both the layer thickness and the respective bulk mean free paths as estimated by Banerjee *et al.*² Consequently, one may argue that there is a discrepancy between the findings of Biagi, Kogan, and Clem¹¹ and the above-described model of a layer-thickness-limited electronic mean free path. However, this conclusion depends crucially on the choice of the values for the Fermi velocities. Note that the above values differ by an order of magnitude from the Fermi velocities one can derive from the system parameters of Banerjee *et al.*² These lead to $v_{F,\text{Nb}} = 0.651 \times 10^7$ cm/s and $v_{F,\text{Cu}} = 0.985 \times 10^7$ cm/s, respectively. When we directly compare the diffusion coefficients of Table I with what Eq. (12) yields a different picture arises. Then the D_{Nb} of Biagi, Kogan, and Clem¹¹ is still smaller than in Ref. 2, but their D_{Cu} is unexpectedly large.

Apart from this somewhat arbitrary choice of Fermi velocities, we have to consider the fact that $H_{c2,\parallel}$ is not included in the analysis. Using the exact solution of the Takahashi-Tachiki equations for the 171.5 Å system with the parameters of Table I, the following findings turn up. First, as a consequence of the extremely large ratio $D_{\text{Cu}}/D_{\text{Nb}}$, the nucleation center of the pair function is situated in the middle of the copper layer over the whole temperature range. Secondly, the dimensional crossover shown by the measurements is not reproduced by the theory. And, finally, at low temperatures the theory underestimates $H_{c2,\parallel}$ by more than a factor of 2, although this may be partly due to the occurrence of surface superconductivity in the samples of Banerjee *et al.*⁴ We must conclude that the parameters do not suffice to give an adequate description of $H_{c2,\parallel}$. More generally, it can be stated that it is easier to fit $H_{c2,\perp}(T)$ than to fit $H_{c2,\parallel}(T)$. The reason for this is that the former shows considerably less structure than the latter. In $H_{c2,\parallel}$ the dimensional crossover has its most dramatic appearance.

TABLE I. Fitting parameters of Biagi, Kogan, and Clem (Ref. 11).

d (Å)	$T_{c,\text{Nb}}$ (K)	D_{Nb} (cm ² /s)	D_{Cu} (cm ² /s)	$N_{\text{Cu}}/N_{\text{Nb}}$	l_{Nb} (Å)	l_{Cu} (Å)
420.5	8.91	2.18	59.1	0.118	24	113
171.5	8.4	1.27	50.2	0.132	14	96

Although Takahashi and Tachiki¹² provided the means for doing advanced calculations, they themselves only applied their equations in an approximate form. This so-called diagonal approximation underestimates H_{c2} and T_c for any real system, but is exact in the thin-layer limit.¹³ In fact, the diagonal approximation is an extension of the Cooper-de Gennes limit to the finite-field case. Still, this approximate form can also reveal some qualitative properties of the upper critical field as a function of temperature, the most profound of these being the dimensional crossover in $H_{c2,\parallel}(T)$. Takahashi and Tachiki¹² demonstrated this by applying it to a set of model systems, mostly ones whose constituent metals differed only in one material parameter. For multilayers differing only in their densities of states a continuous dimensional crossover was found. This crossover was clearly of a different nature than the one found for systems differing only in their diffusion coefficients, e.g., Nb/Nb_{0.6}Ti_{0.4}. In the latter case a discontinuous derivative²³ could be found in $H_{c2,\parallel}(T)$. Karkut *et al.*²⁴ have experimentally affirmed that this anomalous behavior can really occur.

It was claimed¹² that the $H_{c2,\parallel}$ measurements on Nb/Cu multilayers could be well fitted by assuming that both metals differ only in their density of states at the Fermi energy, the ratio $N_{\text{Cu}}/N_{\text{Nb}}$ being equal to 0.15. This compares remarkably well with the value 0.165 that Banerjee *et al.*² estimated from the coefficients of normal-state electronic specific heat. However, the coincidence of the measured and calculated curves may well be fortuitous for reasons that will be discussed in the subsequent analysis. Takahashi and Tachiki¹² do not show absolute quantities. What is plotted is the ratio $H_{c2,\parallel}/H_{c2,\text{Nb}}(0)$ as a function of T/T_c , $H_{c2,\text{Nb}}(0)$ being the upper critical field of bulk niobium at zero temperature and T_c being the multilayer critical temperature. The implication of this is that there are several combinations of system parameters that yield the same curve. These sets can have wildly different values for the diffusion coefficients, which in turn can give rise to wildly different absolute values for $H_{c2,\parallel}$, but leave the ratio $H_{c2,\parallel}/H_{c2,\text{Nb}}(0)$ intact. The reason for this is that both $H_{c2,\parallel}$ and $H_{c2,\text{Nb}}(0)$ are roughly inversely proportional to D , so that the ratio calculated by Takahashi and Tachiki¹² is almost independent of D . Table II shows two combinations of parameters that are consistent with the requirements for the data-fitting curve, namely, $V_{\text{Nb}}=V_{\text{Cu}}$, $D_{\text{Nb}}=D_{\text{Cu}}$, $N_{\text{Cu}}/N_{\text{Nb}}=0.15$, and $d_{\text{Nb}}=d_{\text{Cu}}=0.70\xi_{\text{Nb}}(0)$. Here, $\xi_{\text{Nb}}(0)$ is the Ginzburg-Landau coherence length of niobium at zero temperature, defined by $\xi_{\text{Nb}}(0)=\sqrt{\phi_0/2\pi H_{c2,\text{Nb}}(0)}$. For a bulk superconductor this quantity can be related to the diffusion coefficient and the critical temperature by the formula

$$\xi_{\text{Nb}}(0)=\left[\frac{\hbar D_{\text{Nb}}}{4\pi k T_{c,\text{Nb}}}\exp\left[-\Psi\left(\frac{1}{2}\right)\right]\right]^{1/2}, \quad (13)$$

which was implicitly used by Takahashi and Tachiki. To keep in line with their treatment we did not use the exact frequency summation (10) to calculate the values of Table II, but the large- ω_D approximation¹³ employed by all previous authors. Equation (13) is only valid within this approximation. Remarkably, for both systems there is a nonzero V_{Cu} , due to the requirement $V_{\text{Nb}}=V_{\text{Cu}}$, and consequently a nonzero $T_{c,\text{Cu}}$. However, since N_{Cu} is much smaller than N_{Nb} , this value is extremely small, in the order of 10^{-8} K, and therefore well below the detection limit of any experiment. In the first set of parameters $T_{c,\text{Nb}}$ is set to its bulk value and D_{Nb} is chosen such that we have a layer thickness of $d=0.70\xi_{\text{Nb}}(0)=200$ Å. This is the layer thickness of the sample to which the model system was compared by Takahashi and Tachiki. Applying the diagonal approximation reveals that this set of parameters has a diffusion coefficient of niobium that is much too large, causing this set to yield upper critical fields that underestimate the data by more than a factor of 3. In the second set $T_{c,\text{Nb}}$ and D_{Nb} are adjusted such that the diagonal approximation yields absolute values for T_c and $H_{c2,\parallel}$ that are in quantitative agreement with the data of the same sample. In that case an anomalously high $T_{c,\text{Nb}}$ is needed and the value used for the layer thickness, namely, $d=0.70\xi_{\text{Nb}}(0)$, differs from the sample layer thickness. Moreover, these parameters yield an underestimation of $H_{c2,\perp}$ by a factor of about $\frac{2}{3}$ over the whole temperature range. Therefore the inevitable conclusion arises that Takahashi and Tachiki could only “fit” the data by limiting themselves to reduced quantities, by which important information appears to be obscured. For a real comparison the results should be presented in terms of absolute values, not by using ratios.

Auvil and Ketterson¹⁴ solved the Takahashi-Tachiki equations exactly for the zero-magnetic-field case. They performed a fit to the Nb/Cu T_c data measured by Banerjee *et al.*² by using the niobium T_c as a free parameter. The exact theory yielded a decrease of the niobium critical temperature as a function of niobium layer thickness that was less dramatic than found earlier using Werthamer theory.² Now, the results were in quantitative agreement with the findings of Park and Geballe²⁵ for single niobium films. Both the fit procedure and the system parameters employed in this calculation were copied from Banerjee *et al.*² Therefore this study suffers from the same incompleteness as was discussed in connection with the latter publication. If upper critical fields had been studied as well, it would have come out that the exact theory also cannot adequately describe the reality with this set of parameters.

TABLE II. Parameters of the Takahashi-Tachiki system (Ref. 12).

d (Å)	$T_{c,\text{Nb}}$ (K)	D_{Nb} (cm ² /s)	D_{Cu} (cm ² /s)	$N_{\text{Cu}}/N_{\text{Nb}}$
200	8.91	16.8	16.8	0.15
112	10.7	6.35	6.35	0.15

IV. RESULTS FOR THE V/Ag SYSTEM

We recall that to characterize a specific metal the theory requires the four parameters N , V , D , and Θ_D . The theory does not account for the two metals having different Debye temperatures. Therefore the average

value of 302.5 K is used for both V and Ag, their actual Debye temperatures being 390 and 215 K, respectively. N_{Ag} and V_{Ag} are assumed to be independent of the silver layer thickness. For N_{Ag} this is justifiable, because the Fermi energy is situated in the low, flat *sp* tail of the density of states curve. Therefore neither a shift in the Fermi energy nor a broadening of the density of states can result in a substantial change of the density of states at the Fermi energy. We use $N_{\text{Ag}} = 1.00 \times 10^{47} / \text{J m}^3$, consistent with the measured value of the coefficient of electronic specific heat.⁷ Since silver is not a superconducting metal we assume that $V_{\text{Ag}} = 0$ for all layer thicknesses. From $N_{\text{V(bulk)}}$ and $T_{c,\text{V(bulk)}}$ for *bulk* vanadium (infinite layer thickness) it is possible to determine $V_{\text{V(bulk)}}$. We assume V_{V} to retain this value in vanadium layers of finite thickness, in contrast with N_{V} and $T_{c,\text{V}}$ themselves. This can be justified by realizing that the main influence of V_{V} in the calculations is via the product $N_{\text{V}}V_{\text{V}}$ which fully fixes the vanadium critical temperature. Only small variations of $N_{\text{V}}V_{\text{V}}$ can occur, because $T_{c,\text{V}}$ shows a very strong dependence on this product. This means that a possible layer-thickness dependence of V_{V} can easily be subsumed in the layer-thickness dependence of N_{V} without seriously disturbing the effect of the N -dependent boundary condition. We use $N_{\text{V(bulk)}} = 4.5 \times 10^{47} / \text{J m}^3$ and $T_{c,\text{V(bulk)}} = 4.24$ K. The first value is based on the calculations of Soukoulis and Papaconstantopoulos²⁶ for disordered vanadium, substituting the measured bulk residual resistivity of $15 \mu\Omega \text{ cm}$.⁷ Note that this is significantly lower than the value of $18.3 \times 10^{47} / \text{J m}^3$ that one would derive from the coefficient of electronic specific heat.⁷ The value for $T_{c,\text{V}}$ originates from a measurement on a thick V film.²⁷ The three remaining quantities, N_{V} , D_{V} , and D_{Ag} , are used as free parameters to fit the experimental multilayer T_c and the critical fields $H_{c2,\perp}(T)$ and $H_{c2,\parallel}(T)$ at some temperature T to be chosen. Consequently, these quantities are allowed to vary from system to system and are expected to show a dependence on the thickness of the layers.

The results of the fit procedure applied to the data of Kanoda *et al.*⁷ are shown in Table III. The first two columns show the respective layer thicknesses and the measured T_c of the multilayer system. The third column shows the temperature T at which $H_{c2,\perp}$ and $H_{c2,\parallel}$ are

fitted, always chosen at the lower bound of what was experimentally accessible. The remaining columns show the fitted values for the three free parameters. The calculations were done for infinite multilayers. These can, nevertheless, be compared with measurements in finite samples, because all samples were thick compared to the coherence lengths and protected against surface effects by using Ag layers of at least 100 Å as the outer two layers. The extent to which the experimental error is transferred to the calculated results is estimated and never found to be dramatic. Assuming an error of 1% in T_c , $H_{c2,\perp}(T)$, and $H_{c2,\parallel}(T)$, we find an error of 1% in $T_{c,\text{V}}$ and errors ranging from 2% to 3% in D_{V} and from 2% to 5% in D_{Ag} .

Although the scattering in the results is considerable, the $T_{c,\text{V}}$ we found shows a clear tendency to decrease when the vanadium layers become thinner. This is consistent with the expectation that the density of states is reduced when the mean free path becomes layer-thickness limited. However, in contrast with what was reported for single vanadium films,²⁷ for most of the samples $T_{c,\text{V}}$ exceeds the bulk critical temperature of 4.24 K, V(30 Å)/Cu(15 Å) being the only exception. There seems to be no evidence either experimentally or theoretically that such an enhanced T_c is physical. Most likely, therefore, it should rather be regarded as a deficiency of the theory itself or of the choice of parameters. At the end of this section we will discuss the possibilities to overcome this anomaly.

The diffusion coefficients of both materials also depend on layer thickness, as can be seen in Figs. 2 and 3. The points in the first graph can roughly be grouped onto an ascending straight line, the D_{V} found for V(100 Å)/Ag(200 Å) being an exception to this behavior. But in contrast with the concept of a layer-thickness-limited mean free path, this line would not extrapolate to zero diffusion coefficient for zero layer thickness. For the samples with $d_{\text{Ag}} \leq 400$ Å the silver diffusion coefficient increases with the silver layer thickness, with a slight tendency to saturate at larger thicknesses. The D_{Ag} found for V(240 Å)/Ag(480 Å), however, falls far below the curve that an extrapolation of this tendency would yield.

Using the free-electron formula $D = \frac{1}{3}v_F l$ and the Fermi velocities $v_{F,\text{V}} = 3.73 \times 10^7$ cm/s and $v_{F,\text{Ag}} = 1.39 \times 10^8$ cm/s,⁷ we can calculate the mean free path. For vanadium this quantity ranges from 8 to 36 Å and for silver from 2 to 24 Å. Kanoda *et al.*⁷ estimated a mean free path of 22 Å for vanadium in multilayers with large layer thicknesses. This value falls within the range determined above for vanadium, but the fact that for $d_{\text{V}} > 200$ Å the mean free path is still an increasing function of d_{V} suggests that l_{V} exceeds the value of 200 Å. In the case of silver, the mean free path is always much less than the bulk mean free path, but in all cases also much less than the silver layer thickness. Therefore the procedure introduced by Banerjee *et al.*,² in which the actual mean free path is taken to be the minimum of the bulk mean free path and the respective layer thickness, cannot model the behavior of the silver mean free path. Since these deviant values are needed to fit the data, it seems that the

TABLE III. Fitting parameters for the V/Ag multilayer systems.

$d_{\text{V}}/d_{\text{Ag}}$ (Å/Å)	T_c (K)	T (K)	$T_{c,\text{V}}$ (K)	D_{V} (cm ² /s)	D_{Ag} (cm ² /s)
30/15	2.42	1.6	4.00	1.00	1.07
40/20	2.87	1.6	4.59	1.11	1.48
100/50	3.69	1.6	5.45	1.39	2.97
100/100	3.41	1.6	6.50	1.96	4.54
100/200	2.57	1.6	7.42	4.51	6.97
150/50	4.21	1.5	5.44	1.54	3.07
160/320	2.88	1.5	6.87	2.52	10.7
200/400	3.48	1.5	6.88	1.87	11.1
240/480	3.40	1.6	5.75	2.29	5.46

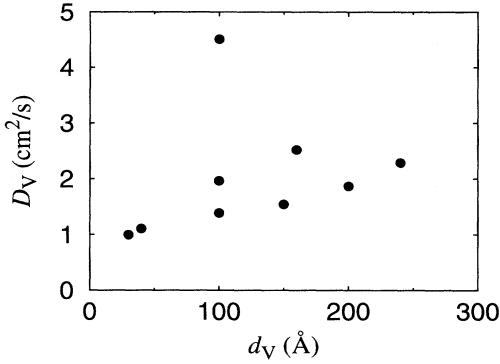


FIG. 2. Layer-thickness dependence of the fitting vanadium diffusion coefficient as listed in Table III.

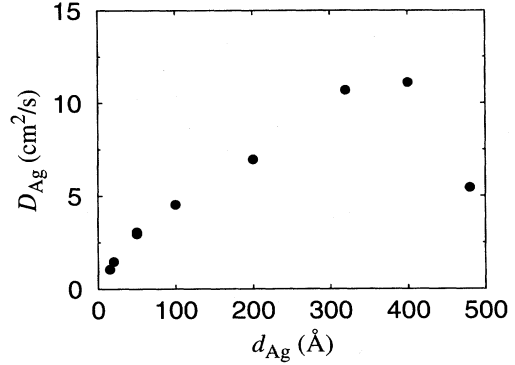


FIG. 3. Layer thickness dependence of the fitting silver diffusion coefficient as listed in Table III.

diffusion coefficient to be used in the Takahashi-Tachiki formalism cannot be directly associated with the diffusion coefficient responsible for the normal-state resistivity. It would, however, also be useful to investigate whether the use of free-electron formulas is justified for the noble and transition metals in the system under consideration.

For the three-dimensional multilayers, i.e., the systems for which no dimensional crossover was observed, the above parameters can reproduce the graphs of Kanoda *et al.*⁷ satisfactorily. In both the calculations and the experiments the anisotropy $H_{c2,\parallel}/H_{c2,\perp}$ is essentially constant over the whole temperature range, meaning that fitting the curve at $T=1.6$ K results in a curve that fits the data at all temperatures. For $H_{c2,\parallel}(T)$ the pair function shows no preference for nucleation in a specific layer. Only at lower temperatures do the systems with $d_V=100$ Å show a slight preference for nucleation in the V layer. Both the constant anisotropy and the lack of preference for the nucleation position are hallmarks of three-dimensional behavior. In these systems the specific properties of V and Ag are averaged out and the multilayer can be regarded as an effective anisotropic bulk material. For V(150 Å)/Ag(50 Å) the experimental data show a discontinuity in the derivative of $H_{c2,\parallel}(T)$ at 2.57 K, attributed to a commensurability between the multilayer period and the vortex lattice. This effect is not reproduced by the Takahashi-Tachiki theory, but otherwise the calculated upper critical fields are in accordance with the measurements. At temperatures below 3 K the theory predicts that the pair function shows a preference for nucleation in the vanadium layer, but the onset of this does not cause an observable upturn of the curve. For V(160 Å)/Ag(320 Å) the experiment shows the onset of a dimensional crossover in $H_{c2,\parallel}(T)$. This is well reproduced by the calculated curve. Again, at lower temperatures nucleation occurs in vanadium. For the two remaining systems, V(200 Å)/Ag(400 Å) and V(240 Å)/Ag(480 Å), the experiments clearly show a dimensional crossover. The calculations show a dimensional crossover too, but the curves do not reproduce the experimental data well. For V(240 Å)/Ag(480 Å) the experimental curve shows the crossover in $H_{c2,\parallel}$ at $T=0.79T_c$. The same crossover, was found to be reflected in $H_{c2,\perp}$ in

terms of a positive curvature. For these experimental curves we refer to Fig. 12 of Ref. 7. Figure 4 shows the corresponding calculated upper critical fields for this system with the parameters of Table III. In this curve $H_{c2,\parallel}$ has its dimensional crossover almost immediately below T_c . Below this crossover point it shows the square-root-like behavior characteristic for decoupled thin films. The perpendicular upper critical field also contrasts with the experimental data by having a negative curvature over the whole temperature range. It is still an open question whether another choice of the fixed parameters can cure this deviation from experiment.

We now return to the problem of the anomalously high vanadium critical temperature. We consider three options to overcome this anomaly. The first is the choice of a lower value for the ratio N_{Ag}/N_V , the second the use of a finite V_{Ag} , and the third the implementation of a finite boundary transparency in the way discussed in Sec. II. For each of these options it can be argued that it increases the multilayer T_c when the other parameters are kept fixed. The effect of both a more extreme N_{Ag}/N_V and a finite boundary resistance R_B is that the behavior of the superconducting layer is shifted towards single-film behavior. In both cases it becomes more difficult for Cooper pairs to diffuse into the normal layers, in the one

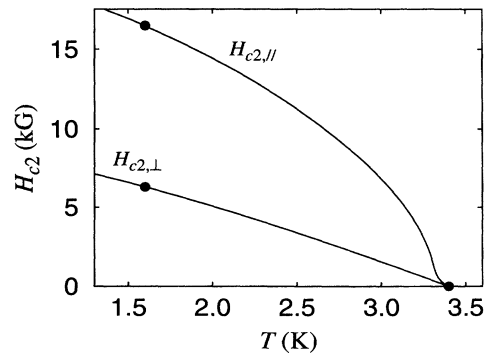


FIG. 4. Upper critical field curve of V(240 Å)/Ag(480 Å). The solid circles show the data points of Kanoda *et al.* (Ref. 7) at which the curves have been fitted.

case due to the lack of available states and in the other due to the difficulty in crossing the interface. Therefore the proximity effect with the nonsuperconducting layer is reduced and a lesser depression of T_c with respect to $T_{c,V}$ is expected. Evidently, when the silver layer is assumed to be a little superconducting by itself, by choosing a finite and positive V_{Ag} , the reduction of T_c induced by the proximity effect with the silver layer is also expected to be less. When one of the above alternatives is used in the fit procedure we presented, this tendency to increase the multilayer T_c has to be compensated by a change in the parameter values $T_{c,V}$, D_V , and D_{Ag} , since the experimental T_c to be fitted obviously remains the same. $T_{c,V}$ can contribute to this compensation by lowering its value. Therefore the above mechanisms are expected to reduce the vanadium T_c that comes out of our fit procedure.

To give a quantitative illustration we consider the V(160 Å)/Ag(320 Å) system. The parameters given in Table III (with $T_{c,V}=6.87$ K) were forced to fit the multilayer T_c of 2.88 K. Merely setting $N_{Ag}/N_{V(bulk)}$ to half its original value and leaving the other parameters unchanged causes the multilayer T_c to increase to 4.41 K. However, again applying the fit procedure causes $T_{c,V}$, D_V , and D_{Ag} to change their values in order to keep the multilayer T_c at 2.88 K. The $T_{c,V}$ we then find is 5.13 K. Similarly, setting V_{Ag} equal to V_V increases the multilayer T_c to 3.30 K, whereas renewed application of the fit procedure yields $T_{c,V}=6.19$ K. In this case we have $N_{Ag}V_{Ag}=0.050$. This compares nicely to the data of Deutscher *et al.*,²⁸ who reported that the silver layer in a PbBi/Ag/PbBi sandwich can become intrinsically superconducting due to the proximity of the superconductor. The silver T_c associated with this value of $N_{Ag}V_{Ag}$ is 7.33×10^{-7} K. Finally, the introduction of a finite boundary resistance $R_B=390 \times 10^{-8} \mu\Omega \text{ cm}^2$ gives a multilayer T_c of 3.05 K, provided that the other parameters remain unchanged. It results in a vanadium T_c of 6.66 K when the fit procedure is again applied. All these modifications do not lead to qualitative changes in the behavior $T_{c,V}$, D_V , and D_{Ag} as a function of layer thickness, nor in the behavior of $H_{c2,\parallel}$ and $H_{c2,\perp}$ as functions of temperature. It can be concluded that the tendencies are all in the right direction, but they do not suffice to eliminate the problem of the high $T_{c,V}$.

In order to get a feeling for the measure of the influence of a boundary resistance, it is convenient to express this quantity in terms of a characteristic length scale. That enables one to compare the magnitude of R_B to the coherence lengths and layer thicknesses. The dimension of boundary resistance is $\mu\Omega \text{ cm}^2$. Therefore an appropriate way to attribute a length scale to the boundary resistance is by dividing it by the resistivity of either vanadium or silver. For the V(160 Å)/Ag(320 Å) system with $R_B=390 \times 10^{-8} \mu\Omega \text{ cm}^2$, we find $R_B/\rho_V=12.1$ Å and $R_B/\rho_{Ag}=11.4$ Å. The interpretation of the latter value is that an interface with a certain area A has the same resistance as a 11.4 Å silver layer with the same cross section A .

In connection with the suggestion of a finite R_B , it is also important to consider its influence on the anisotropy

ratio $H_{c2,\parallel}/H_{c2,\perp}$. In thin-layer systems and for $R_B=0$, this ratio is a function of

$$\gamma = \frac{1}{2}(N_{Ag}D_{Ag}/N_V D_V + N_V D_V/N_{Ag}D_{Ag})$$

only.¹³ The minimum value of γ is 1, corresponding to no anisotropy, that is, $H_{c2,\parallel}=H_{c2,\perp}$. A finite boundary resistance tends to increase the anisotropy, meaning that there is also anisotropy for $\gamma=1$. Consequently, R_B imposes a lower bound on the anisotropy one can achieve theoretically by varying the diffusion coefficients. When this minimum value exceeds the experimentally observed anisotropy, the data cannot be fitted any more. For the V(30 Å)/Ag(15 Å) system this happens for $R_B > 1440 \times 10^{-8} \mu\Omega \text{ cm}^2$. Therefore the boundary resistance cannot be increased without restriction and is limited as a means of reducing the vanadium critical temperature.

Another fundamental point to be dwelled upon in connection with the employed fit procedure is the fact that the solutions are not unique. This can be seen most clearly by considering the thin-layer limit. Here, the only way in which the diffusion coefficients appear in the expressions for $H_{c2,\parallel}$ and $H_{c2,\perp}$ is by means of the products $N_V D_V$ and $N_{Ag} D_{Ag}$.¹³ Moreover, all expressions are symmetric with respect to the exchange of these products. As a consequence, in the thin layer, the above-described fit procedure makes it possible to determine the best fitting value for quantities like γ , but leaves it impossible to decide whether $N_V D_V$ is larger than $N_{Ag} D_{Ag}$ or vice versa. Therefore two possible solutions are left to choose from for each system. Farther away from the thin-layer limit the situation is the same in the sense that two solutions still exist, although these need not correspond to equal values of γ . In the present work we have always chosen the solution with the highest value for the ratio $N_V D_V/N_{Ag} D_{Ag}$. For this choice the initial circumstance that $N_V > N_{Ag}$ is slightly counterbalanced by the fact that a $D_V < D_{Ag}$ is found. This leads to moderate values for the diffusion coefficients, which are not too far off from the prediction of Eq. (12). The fact that this implies for most systems that $\rho_V < \rho_{Ag}$ is understandable from the notion that bulk silver derives its small resistance from its large mean free path. In multilayers this does not apply anymore, because the mean free path has become layer-thickness limited. The second solution corresponds to $N_V D_V/N_{Ag} D_{Ag}$ well below 1, meaning that the contribution of the densities of states to this ratio has to be more than completely balanced by the diffusion coefficients. This results in both an extremely low D_V and an extraordinary high D_{Ag} , when compared to the prediction of Eq. (12). To illustrate this, consider the V(30 Å)/Ag(15 Å) system. The ratio $N_V D_V/N_{Ag} D_{Ag}$ 4.16 and 0.230 for the first and second solution, respectively. These values correspond to approximately the same γ value, as the layers are comparatively thin. But the corresponding values for D_{Ag}/D_V are 1.07 and 19.3, respectively, differing by an order of magnitude. Similarly, for the V(200 Å)/Ag(400 Å) system we have for $N_V D_V/N_{Ag} D_{Ag}$ the values of 0.844 and 0.0299 for the first and the second solution, this system being the only

example in Table III for which the first yields a number less than unity. Here, the corresponding values for $D_{\text{Ag}}/D_{\text{V}}$ are, respectively, 5.94 and 163, the latter being extremely large. In calculating $H_{c2,\parallel}(T)$, the high $D_{\text{Ag}}/D_{\text{V}}$ value of the second solution leads to the seemingly unphysical, but theoretically possible effect that the pair function can also nucleate in the middle of the silver layer, and will indeed do so above the specific crossover temperature of 1.89 K. Below this temperature nucleation occurs in the middle of the vanadium layer, whereas at this temperature $H_{c2,\parallel}$ has a discontinuity in its temperature derivative. Since the experiments of Kanoda *et al.*⁷ show no sign of the existence of such a discontinuity, the choice for the first solution seems to be the most reasonable one. In a previous publication²⁹ the present authors have presented results that correspond to the second possible solution. These results suffered from the same high critical temperatures of vanadium that are encountered above. It was found that for thick-layer systems the calculated $H_{c2,\parallel}(T)$ curve underestimated the experiment everywhere between T_c and the fitting temperature. We presently believe that this choice for the second solution is not the best option available.

Taking things together, there are two main problems in fitting the experimental data. First, unphysically high values for the critical temperature of vanadium are found and, secondly, the dimensional crossover in $H_{c2,\parallel}$ is found at higher temperatures than observed experimentally. The first problem can be cured by decreasing $N_{\text{Ag}}/N_{\text{V}}$ or increasing V_{Ag} , but such a choice for the material parameters lacks an external justification. The problem cannot be overcome by introducing a boundary resistance, because the values then needed for the thick-layer systems make it impossible to fit the thin-layer systems. The second problem, concerning the crossover temperature, or, stated differently, concerning the fitting of $H_{c2,\parallel}$ at all temperatures, seems to be inherent to the theory used.

V. RESULTS FOR THE Nb/Cu SYSTEM

For Nb/Cu the same procedure is adopted as used for V/Ag. We use a Debye temperature of 295 K, which is the average of the Debye temperatures of pure Nb and Cu, 275 and 315 K, respectively. The other fixed input parameters, taken from Banerjee *et al.*,² are, respectively $N_{\text{Cu}} = 1.98 \times 10^{47} / \text{J m}^3$, $V_{\text{Cu}} = 0$, $N_{\text{Nb(bulk)}} = 12.0 \times 10^{47} / \text{J m}^3$, and $T_{c,\text{Nb(bulk)}} = 8.91$ K. From the latter two values V_{Nb} is determined, which is again assumed to be layer-thickness independent. Now N_{Nb} , D_{Nb} , and D_{Cu} are the free parameters, used to fit the multilayer T_c and the critical fields $H_{c2,\perp}(T)$ and $H_{c2,\parallel}(T)$ for some temperature T to be chosen.

The above fit procedure has been applied to the data of Chun *et al.*⁵ In their experiments surface superconductivity was suppressed by sandwiching the necessarily finite multilayers between thick copper layers. Since all samples were thick with respect to the coherence length, this made the behavior of their samples also representative for what an infinite multilayer would do. Therefore the data can be compared to our calculations for infinite multilayers. The resulting values for the free parameters

are listed in Table IV. The first three columns show the layer thicknesses, the multilayer T_c , and the temperature at which $H_{c2,\perp}$ and $H_{c2,\parallel}$ are fitted. The fourth and the last two columns show the results of the above fit procedure. The fifth column shows the niobium T_c as resulting from the fit procedure proposed by Auvil and Ketterson.¹⁴ Here only the multilayer T_c is fitted using $T_{c,\text{Nb}}$ as the free parameter and keeping V_{Nb} equal to its bulk value. The diffusion coefficients used for this fit are determined from Eq. (12) of Ref. 2. Assuming an experimental error of 1% for the measured temperatures and fields, we find for fits with $T/T_c < 0.67$ an error of 1% in $T_{c,\text{Nb}}$ and errors ranging from 2% to 3% in D_{Nb} and from 2% to 5% in D_{Cu} . For fits with T closer to T_c the relative error in the diffusion coefficients is approximately equal to the relative error in $T_c - T$, due to the fact that the diffusion coefficients are roughly inversely proportional to dH_{c2}/dT at T_c . Since this error is $T_c/(T_c - T)$ times the error in T_c , it will always become large for T lying close to T_c . Two of the measurements listed by Chun *et al.*⁵ are not included in Table IV, namely, Nb(176 Å)/Cu(585 Å) at $T = 1.10$ K and Nb(175 Å)/Cu(1240 Å) at $T = 1.17$ K. For these measurements no parameters could be found that reproduced the data, meaning that they have to be regarded as inconsistent with the present theory combined with the choice of the fixed parameters.

Figure 5 shows the values of $T_{c,\text{Nb}}$ as a function of d_{Nb} .

TABLE IV. Fitting parameters for the Nb/Cu multilayer systems.

$d_{\text{Nb}}/d_{\text{Cu}}$ (Å/Å)	T_c (K)	T (K)	$T_{c,\text{Nb}}$ (K)	$T_{c,\text{Nb}}^{\text{Auvil}}$ (K)	D_{Nb} (cm ² /s)	D_{Cu} (cm ² /s)
23/23	3.34	1.17	6.79	6.56	2.29	2.22
47/22	6.05	1.17	8.24	8.08	2.32	2.50
44/44	5.07	1.17	9.23	8.77	3.65	3.84
47/87	3.05	1.17	9.04	7.96	7.94	10.10
168/147	7.00	1.37	10.14	9.88	3.49	7.26
168/147	7.00	3.33	10.39	9.88	3.50	11.75
168/147	7.00	4.86	10.42	9.88	3.82	11.62
168/147	7.00	5.28	10.39	9.88	3.90	10.81
168/147	7.00	6.04	10.30	9.88	4.31	8.79
168/147	7.00	6.62	10.28	9.88	4.95	8.05
171/376	5.85	1.17	10.15	10.75	4.69	7.76
171/376	5.85	3.04	11.20	10.75	5.12	14.44
171/376	5.85	3.63	11.76	10.75	4.87	20.94
171/376	5.85	4.08	11.92	10.75	5.39	22.22
171/376	5.85	4.47	11.87	10.75	7.04	19.77
171/376	5.85	4.86	11.72	10.75	8.91	17.13
171/376	5.85	5.28	11.47	10.75	11.34	14.34
171/376	5.85	5.41	11.47	10.75	10.36	14.55
171/376	5.85	5.62	11.33	10.75	12.12	13.22
170/90	7.70	1.17	9.80	9.77	3.21	3.29
168/168	6.75	1.17	10.03	9.87	4.07	6.52
168/168	6.75	1.96	10.21	9.87	4.07	8.36
168/168	6.75	5.28	10.33	9.87	5.31	9.03
172/255	6.50	1.56	10.73	10.46	4.10	10.07
172/333	6.05	1.10	10.14	10.62	3.56	7.68
172/333	6.05	1.17	10.04	10.62	3.77	7.04
176/585	4.84	3.33	9.91	10.62	5.87	11.62
176/585	4.84	3.77	11.51	10.62	5.31	25.23

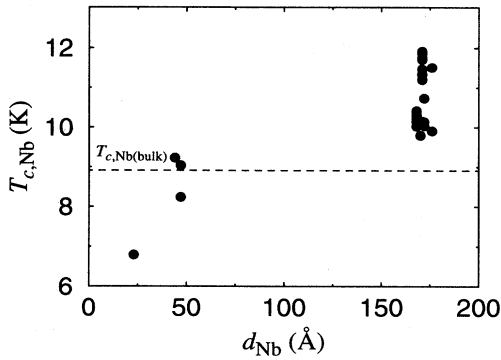


FIG. 5. Layer-thickness dependence of the fitting niobium critical temperatures as listed in Table IV.

As for vanadium in V/Ag, it is observed that lower values for $T_{c,Nb}$ are needed when d_{Nb} decreases. However, for most samples this value is found to exceed the bulk critical temperature. This contrasts with the behavior of single niobium films,^{22,25} for which T_c is always below the bulk T_c . The temperatures arising from the fit procedure of Auvil and Ketterson show the same anomaly, though in general their scheme leads to a slightly lower $T_{c,Nb}$ than the full fit with varying diffusion coefficients. The exceptions to this rule are precisely those systems for which the fitted D_{Cu} in Table IV is below the value of Eq. (12). Since a smaller D_{Cu} diminishes the extension of the pair function into the copper layer, due to a shorter coherence length,¹³ it tends to decrease the proximity-effect coupling, thereby enhancing the multilayer T_c . Consequently, for these systems the full fit procedure leads to a smaller $T_{c,Nb}$ to compensate this effect of a smaller D_{Cu} . The fact that the use of Auvil and Ketterson's fit procedure also leads to high niobium critical temperatures contrasts with the findings of Auvil and Ketterson¹⁴ themselves. Using the same procedure, but applying it to another set of data,² they found niobium critical temperatures that were in agreement with the single-film behavior reported by Park and Geballe.²⁵

The layer-thickness dependence of the niobium

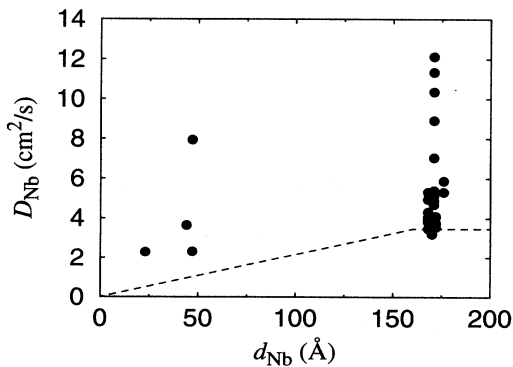


FIG. 6. Diffusion coefficient of niobium as a function of layer thickness. The solid circles are the result of our fit procedure, as listed in Table IV. The dashed line shows the value according to Eq. (12).

diffusion coefficient is shown in Fig. 6. Clearly, the thin-layer values for D_{Nb} cannot well be extrapolated to large layer thicknesses. It is difficult to observe any tendencies, because the points are not evenly distributed along the horizontal axis. For most of the fits with $d_{Nb} \approx 170$ Å the values found for D_{Nb} lie close together in the interval between 3.2 and 5.9 cm^2/s . The points that fall above this cluster of points originate from the Nb(171 Å)/Cu(376 Å) system only. For the thin-layer systems one expects reduced values for the diffusion coefficient, due to the constraint of the mean free path by the thickness of the layer. This effect is indeed observed for three of the four thin-layer systems, but Nb(47 Å)/Cu(87 Å) has resulted in a value that is too high to support this idea. The dashed line shows the diffusion coefficient according to Eq. (12). It is found that application of the fit procedure leads in general to higher values than application of Eq. (12).

In Fig. 7 the copper diffusion coefficient is seen to show the tendency to increase with increasing layer thickness. However, also in this case the picture is somewhat obscured by the large scattering in the data points. No saturation effect can be observed at larger layer thicknesses, as is consistent with the expected large mean free path of the bulk copper system. As for niobium, most of the values found are higher than the prediction of Eq. (12), again represented by the dashed line.

The Fermi velocities that Biagi, Kogan, and Clem¹¹ used in the analysis of their $H_{c2,1}$ fits were $v_{F,Nb} = 2.73 \times 10^7$ cm/s and $v_{F,Cu} = 1.57 \times 10^8$ cm/s. When calculating the electron mean free paths with these numbers we find values ranging from 25 to 133 Å for niobium and from 4 to 48 Å for copper. These values are always far below both the layer thicknesses and the bulk mean free paths, the latter, according to Banerjee *et al.*,² being 160 and 2000 Å, respectively. However, these low numbers originate from the Fermi velocities employed. When using the values that were derived from the system parameters of Banerjee *et al.*,² quite another result is found. Then all the data points in Figs. 6 and 7 that lie above the rising part of the dotted straight line (or its extrapolation to larger thicknesses) correspond to mean free paths that are larger than the layer thickness.

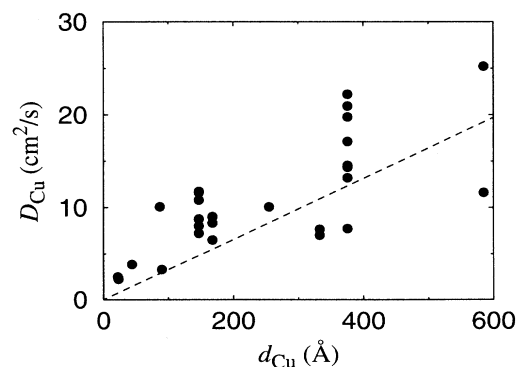


FIG. 7. Diffusion coefficient of copper as a function of layer thickness. The solid circles are the result of our fit procedure, as listed in Table IV. The dashed line shows the value according to Eq. (12).

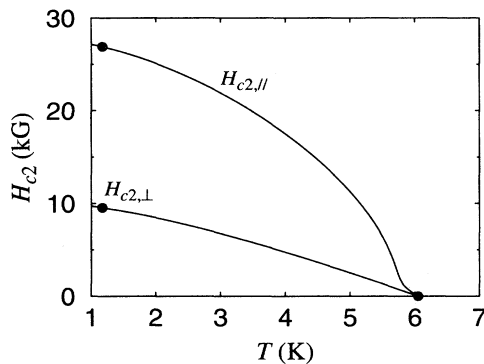


FIG. 8. Upper critical field curve of Nb(172 Å)/Cu(333 Å). The solid circles show the data points of Chun *et al.* (Ref. 5) at which the curves have been fitted.

Figure 8 shows the calculated phase diagram of a representative system with a dimensional crossover in $H_{c2,\parallel}(T)$. The crossover is seen to occur immediately below T_c , at 5.8 K. At the high-temperature side of this point the nucleation center has no preference for a specific layer. At the low-temperature side nucleation takes place in the Nb layer. In the latter region the curve shows the square-root-like behavior characteristic for the two-dimensional regime. Here, there is no coupling between two neighboring niobium layers and there is only the proximity effect of the nucleation layer with the adjacent copper layers. Experimentally (see Fig. 3 of Ref. 5) the dimensional crossover of this system is observed at 3.3 K, that is, at a much lower temperature. This causes the calculation to yield an overestimation of the measured critical field in the region between the fitting temperature of 1.17 K and the critical temperature of 6.05 K. As can be seen, the diagram looks very much the same as the V/Ag result depicted in Fig. 4. Also the deviations from the measured curve⁵ have the same character for both systems.

For Nb/Cu the high critical temperature of niobium can be restricted in the same fashion as described for V/Ag. Reducing $N_{\text{Cu}}/N_{\text{Nb(bulk)}}$ to 0.1, the niobium T_c , we find for the Nb(172 Å)/Cu(333 Å) system (with fitting temperature 1.17 K) is 8.83 K, whereas the original ratio gave 10.04 K. By setting $V_{\text{Cu}} = V_{\text{Nb}}$ we achieve a reduction from 10.04 to 9.56 K. This change would imply a bulk copper critical temperature of 5.76×10^{-8} K. Introducing a boundary resistance $R_B = 390 \times 10^{-8} \mu\Omega \text{ cm}^2$ brings us from 10.04 to 9.74 K. The characteristic lengths for this resistance are $R_B/\rho_{\text{Nb}} = 44.1 \text{ \AA}$ and $R_B/\rho_{\text{Cu}} = 17.1 \text{ \AA}$. The boundary resistance at which it becomes impossible to fit, for example, the Nb(23 Å)/Cu(23 Å) system is $612 \times 10^{-8} \mu\Omega \text{ cm}^2$.

The same conclusions can be drawn as for the V/Ag multilayer. We find anomalously high critical temperatures for niobium and higher dimensional crossover tem-

peratures than experimentally observed. Decreasing $N_{\text{Cu}}/N_{\text{Nb}}$ or increasing V_{Cu} can relieve the first anomaly, but the problem cannot be solved by using a finite boundary resistance. The lack of coincidence of the measured and calculated phase diagrams for a parallel magnetic field seems to be deeply rooted in the combination of parameters and theory employed. The only known escape would be to use, while performing the calculations, a "theoretical" layer thickness smaller than the actual one. This has been done effectively by Takahashi and Tachiki,¹² as we have seen above in Sec. III, Table II, but at present a justification is lacking for such a route.

VI. CONCLUSIONS

We have presented results of calculations in which the Takahashi-Tachiki theory is applied without approximations. We have compared these results with experimental data for V/Ag and Nb/Cu. To our knowledge, this is the first time that the exact solution of the Takahashi-Tachiki equations at finite fields has been compared with experimental data. We used the critical temperature of the superconducting metal and the diffusion coefficients of both metals as free parameters to fit the experimental data. For 3D systems the phase diagrams could be well reproduced. However, for 2D systems difficulties were encountered. For these, the parameters that fitted the data at best showed an anomalous behavior. For most fitted samples the critical temperatures of both V and Nb were found to be larger than the bulk T_c and thus did not obey the expected single-film behavior. The diffusion coefficients of V, Ag, Nb, and Cu were much lower than one would expect from resistivity measurements, but this conclusion strongly depends on the Fermi velocities assumed for these materials. The diffusion coefficients of Nb and Cu were not in contradiction with the model that Banerjee *et al.* introduced, but the scattering in the data points was too large to regard the calculations as a confirmation. The theoretical dimensional crossover in the parallel upper critical field could not well be made to coincide with the experimental one. The results throw a new light on the findings of Auvil and Ketterson. Their agreement of the Nb critical temperature with single-film behavior was obtained with a set of parameters that cannot well describe the upper critical fields. The complete analysis of the absolute values of upper critical fields has also revealed the fortuity of a suggestive figure in which Takahashi and Tachiki fitted a model calculation with $D_{\text{Nb}} = D_{\text{Cu}}$ and $V_{\text{Nb}} = V_{\text{Cu}}$ to Nb/Cu data.

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