Perpendicular upper critical field and critical temperature of superconducting–interphase–normal-metal multilayers

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Proximity-effect theory of superconductor–normal-metal (S/N) superlattices is extended to the case of multilayers with an interphase between S and N layers. A nonmonotonic dependence of the superlattice critical temperature on the period Λ is obtained theoretically, in accordance with experiments of Qian *et al.* on Nb/Ti. Similar behavior of the perpendicular upper critical field H_{c2} is predicted.

I. INTRODUCTION

Artificially layered superconducting superlattices recently have raised considerable interest, because they can have properties that differ significantly from those of the constituent bulk metals. The critical temperatures, fields, and currents all show very interesting dependences on the superlattice period.

Several attempts to describe these systems theoretically have been made using the de Gennes–Werthamer proximity-effect theory[1] for dirty-superconductor–normal-metal (S/N) bilayers. However, the agreement between the theory and experimental data for critical temperatures and fields is in many cases only qualitative. One interesting situation where a generalization of the simple bilayer theory is obviously needed, and which is often met in practice, is the case when an interphase (I) is formed between the two metals constituting the superlattice. While in the two-metal (S/N) superlattice the de Gennes–Werthamer theory predicts (Refs. 1 and 2) a monotonic dependence of T_c on the superlattice period, the presence of an interphase can cause nonmonotonicity. In the present paper we extend the theory of Biagi, Kogan, and Clem[3] for S/N superlattices to this case to calculate the perpendicular upper critical field H_{c2} and the critical temperature T_c. We actually treat the general case for which all three layers (S, I, and N) are superconductors, but the transition temperature of N is lower than that of S.

We demonstrate the possibility of explaining by our theory the experimentally observed[4] increase of T_c at small superlattice periods, where the interphase effect becomes important. We predict similar effects in the behavior of H_{c2}. We also show that our main equation for the superlattice made of two metals with the interphase between them gives the same critical temperature as that obtained by Triscone *et al.*[5] for a single trilayer.

The organization of this paper is as follows: In Sec. II we give a system of equations for the critical temperature and the perpendicular upper critical field of a superconductor-interphase–normal-metal ($S/I/N$) superlattice. In Sec. III we compare our equations with the equations for a single $S/I/N$ trilayer.[5] We compare theory and experiment in Sec. IV and give a summary in Sec. V.

II. DERIVATION OF THE MAIN EQUATION

We consider an infinite stack of alternating S and N layers (parallel to the x-y plane) with an I layer between two neighboring N and S layers (Fig. 1). The period of the superlattice is $\Lambda = d_S + d_N + 2d_I$, where $d_{S,N,I}$ are the thicknesses of S, N, and I layers.

Following Ref. 3 we use the quasiclassical equations of superconductivity in the dirty limit[6]

$$-\frac{D}{2}\mathbf{\Pi}\cdot(G\mathbf{\Pi}F - F\mathbf{\Pi}G) = \frac{\Delta}{\hbar}G - \omega F , \quad (1)$$

and

$$G^2 + |F|^2 = 1 , \quad (2)$$

where D is the diffusion coefficient, $\mathbf{\Pi} = \nabla - (2\pi i/\phi_0)\mathbf{A}$, and $\mathbf{A}$ is the vector potential.

Near the phase transition to the normal state, we seek the solutions for $G(\mathbf{r},\omega)$ and $F(\mathbf{r},\omega)$, the Gorkov Green's functions integrated over the energy and averaged over the Fermi surface, in the form

$$G \approx 1, \quad F(\mathbf{r}) = \frac{\Delta(\mathbf{r})}{\hbar\omega + 2\pi k_B T_{ci}\,\rho(T/T_{ci})} \quad (3)$$

in each S, N, or I region. Here $\Delta(\mathbf{r})$ is the order parameter, T_{ci} is the critical temperature of the bulk i metal ($i = S$, N, or I, i.e., $T_{ci} = T_{cS}$, T_{cN}, or T_{cI}) and $\omega = \pi k_B T(2n+1)/\hbar$ is the Matsubara frequency with $n = 0, 1, 2, \ldots$. The self-consistency relation, which relates F and Δ, defines the functions $\rho = \rho(t_i)$ via

$$\ln(t_i) = \Psi(\tfrac{1}{2}) - \Psi\left[\frac{1}{2} + \frac{\rho}{t_i}\right] , \quad (4)$$

where $t_i = T/T_{ci}$ is the reduced temperature and Ψ is the digamma function. In each region $F(\mathbf{r})$ is governed by

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an equation of the form

$$\Pi^2 F = -k^2 F \ . \tag{5}$$

Assuming the external field in the direction transverse to the layers, $\mathbf{H}=H\hat{\mathbf{z}}$, we choose the gauge $\mathbf{A}=(0,Hx,0)$ and separate variables in Eq. (5):

$$F(\mathbf{r})=f(x,y)g(z) \ . \tag{6}$$

This gives

$$-(\Pi_x^2+\Pi_y^2)f_{S,I}(x,y)=(k_{S,I}^2-q_{S,I}^2)f_{S,I}(x,y) \ , \tag{7a}$$

$$\frac{d^2 g_{S,I}}{dz^2}=-q_{S,I}^2 g_{S,I}(z) \ , \tag{7b}$$

for S and I layers and

$$-(\Pi_x^2+\Pi_y^2)f_N(x,y)=(-k_N^2+q_N^2)f_N(x,y) \ , \tag{7c}$$

$$\frac{d^2 g_N}{dz^2}=q_N^2 g_N(z) \ , \tag{7d}$$

for N layers. Here

$$k_{S,I}^2=\frac{2\rho(t_{S,I})}{\xi_{S,I}^2} \tag{8a}$$

and

$$k_N^2=-\frac{2\rho(t_N)}{\xi_N^2} \ , \tag{8b}$$

where

$$\xi_i^2=\frac{\hbar D_i}{2\pi k_B T_{ci}} \quad (i=S,N,I) \tag{9}$$

is proportional to the corresponding diffusion coefficient D_S, D_N, or D_I. Assuming that the solutions of Eqs. (7) are related at the interfaces by the same boundary conditions as in Ref. 3, we take

$$f_S=f_I, \quad \frac{d}{dz}\ln g_S=\eta_S\frac{d}{dz}\ln g_I \ , \tag{10}$$

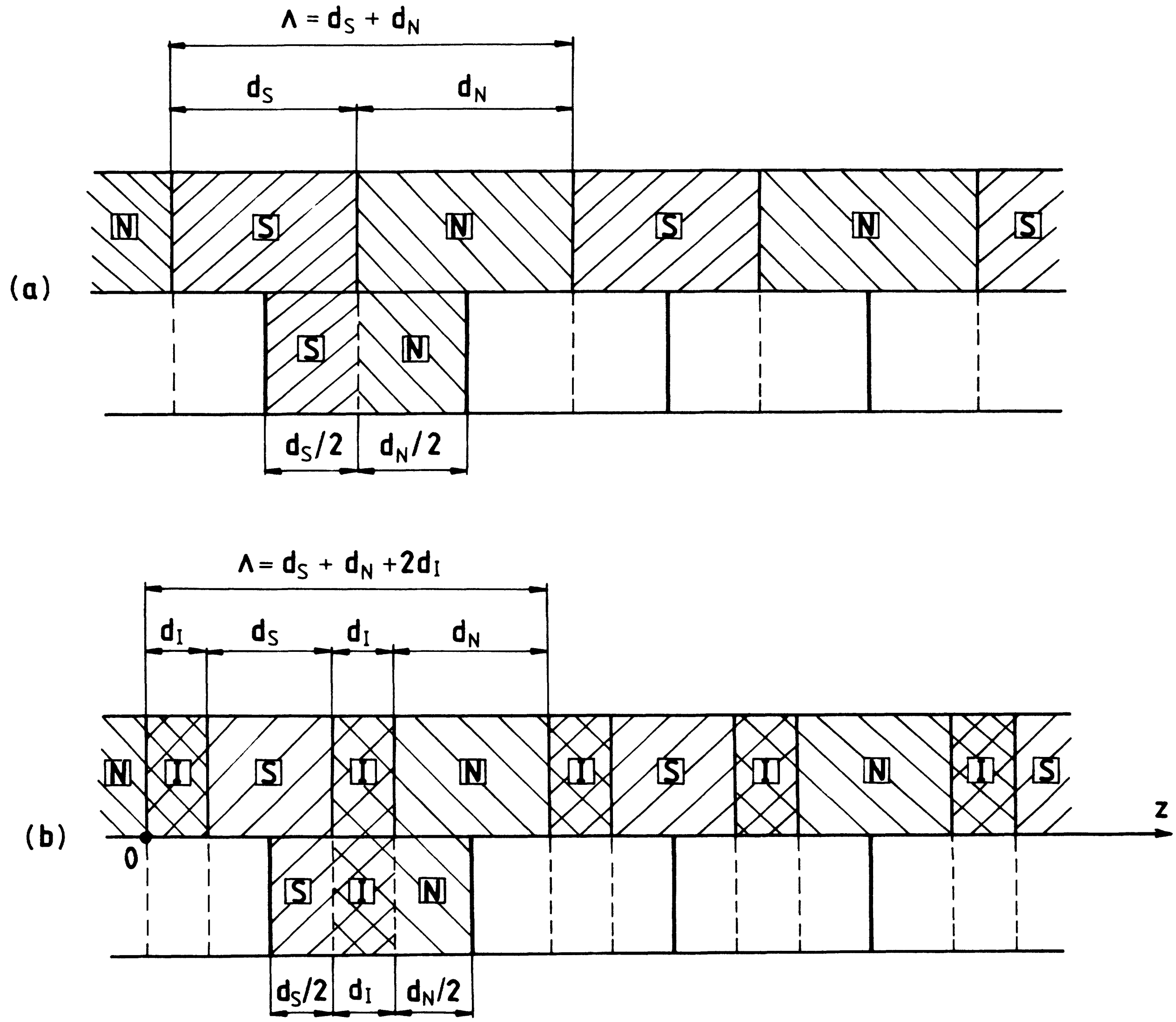

FIG. 1. (a) Two-metal S/N superlattice and its decomposition to corresponding half-thickness bilayers. (b) $S/I/N$ superlattice with interphases I between neighboring S and N layers and its decomposition to corresponding trilayers (see text).

at the S/I interface, and

$$f_N = f_I, \quad \frac{d}{dz}\ln g_N = \eta_N \frac{d}{dz}\ln g_I \tag{11}$$

at the N/I interface. The parameters η_S and η_N characterize the interfaces [$\eta_{S,N} = \sigma_I/\sigma_{S,N}$ in the dirty limit for specular scattering, σ_i ($i = S,N,I$) being the normal state conductivities].

Similar to the case of bimetallic superlattices, one can construct a solution $F(\mathbf{r})$ corresponding to the lowest eigenvalue of Eq. (7) and satisfying the Bloch condition along the z axis. It describes the nucleation of vortices threading through the multilayer.

The corresponding critical field H_{c2} is obtained, using the boundary conditions (10) and (11), from the condition for the existence of a nontrivial solution (see Appendix):

$$\begin{aligned}1 &= \cos(q_S d_S)\cosh(q_N d_N)\cos(2q_I d_I) \\ &+ \frac{1}{2}\left[\frac{q_I^2}{q_S q_N}\eta_S\eta_N - \frac{1}{\eta_S\eta_N}\frac{q_S q_N}{q_I^2}\right]\sin(q_S d_S)\sinh(q_N d_N)\sin^2(q_I d_I) \\ &+ \frac{1}{2}\left[\frac{q_N}{q_I}\frac{1}{\eta_N} - \frac{q_I}{q_N}\eta_N\right]\cos(q_S d_S)\sinh(q_N d_N)\sin(2q_I d_I) \\ &+ \frac{1}{2}\left[\frac{q_N}{q_S}\frac{\eta_S}{\eta_N} - \frac{\eta_N}{\eta_S}\frac{q_S}{q_N}\right]\sin(q_S d_S)\sinh(q_N d_N)\cos^2(q_I d_I) \\ &- \frac{1}{2}\left[\frac{q_S}{q_I}\frac{1}{\eta_S} + \frac{q_I}{q_S}\eta_S\right]\sin(q_S d_S)\cosh(q_N d_N)\sin(2q_I d_I)\ ,\end{aligned} \tag{12}$$

where

$$q_{S,I}^2 = k_{S,I}^2 - \frac{2\pi H_{c2}}{\phi_0}, \quad q_N^2 = k_N^2 + \frac{2\pi H_{c2}}{\phi_0}\ . \tag{13}$$

When combined with Eqs. (4) and (8), Eqs. (12) and (13) give $H_{c2}(T)$ as the highest possible value of the field at a given temperature T. The superlattice critical temperature T_c is obtained for $H_{c2}=0$.

III. MULTILAYER VERSUS TRILAYER PROBLEM

In general, a superlattice can be made of subsequent n layers periodically repeated with $n = 2,3,4,\ldots$. For $n=2$, a bimetallic S/N superlattice, it is easy to show that Eq. (12) with $d_I = 0$ is equivalent to a simple de Gennes–Werthamer relation for a bilayer,[1] with $d_S \to d_S/2$ and $d_N \to d_N/2$. Assuming $dg/dz = 0$ at the outer boundaries of the bilayer [see Fig. 1(a)] and applying the usual boundary conditions at the S/N interface, one gets[3]

$$q_S \tan\frac{q_S d_S}{2} = \eta q_N \tanh\frac{q_N d_N}{2}\ , \tag{14}$$

with $\eta = \sigma_N/\sigma_S$. The equivalence of Eqs. (12) with $d_I = 0$ and (14), due to the symmetry of the problem, can be proved by simple trigonometric transformations.

The same symmetry-based arguments were used by Triscone *et al.*[5] to reduce the $S/I/N$ multilayer problem to that of a trilayer [see Fig. 1(b)] in order to calculate the critical temperature. Replacing in their result k_i by q_i ($i = S$, N, or I), one gets H_{c2} from

$$q_S \tan\frac{q_S d_S}{2} = \eta_S q_I \frac{\alpha - \tan(q_I d_I)}{1 + \alpha\tan(q_I d_I)}\ , \tag{15}$$

with

$$\alpha = \frac{q_N}{\eta_N q_I}\tanh\frac{q_N d_N}{2}\ .$$

To show that this equation yields the highest superlattice T_c and H_{c2}, we note that, assuming

$$\cos(q_S d_S/2)\cos(q_I d_I)\cosh(q_N d_N/2) \neq 0\ ,$$

Eq. (12) can be put into the form

$$Q_1 Q_2 = 0\ , \tag{16a}$$

where

$$\begin{aligned}Q_1 &= \frac{q_S}{q_I}\frac{1}{\eta_S}\tan\frac{q_S d_S}{2} + \tan(q_I d_I) - \frac{q_N}{q_I}\frac{1}{\eta_N}\tanh\frac{q_N d_N}{2} \\ &+ \frac{1}{\eta_S\eta_N}\frac{q_S q_N}{q_I^2}\tan\frac{q_S d_S}{2}\tan(q_I d_I)\tanh\frac{q_N d_N}{2}\ ,\end{aligned} \tag{16b}$$

and

$$\begin{aligned}Q_2 &= -\frac{q_I}{q_S}\eta_S\tan\frac{q_S d_S}{2} - \tan(q_I d_I) - \frac{q_I}{q_N}\eta_N\tanh\frac{q_N d_N}{2} \\ &+ \frac{q_I^2}{q_S q_N}\eta_S\eta_N\tan\frac{q_S d_S}{2}\tan(q_I d_I)\tanh\frac{q_N d_N}{2}\ .\end{aligned} \tag{16c}$$

Equation (16a) means that $Q_1 = 0$ or $Q_2 = 0$. However, the condition $Q_1 = 0$ is exactly Eq. (15) for a trilayer. This means, as Triscone *et al.*[5] pointed out, that the trilayer T_c gives the lower bound of T_c for a superlattice with interphase layers. Also, for $q_S d_S/2 < \pi/2$ (which is usually the case, except in proximity with magnetic metal[7]) and $q_I d_I \ll 1$, Q_2 is always negative so that the trilayer model gives the same result as the superlattice calculation. The same conclusion holds if $q_I^2 < 0$ (which, if we want to find T_c, means that $T_{cI} < T_c$). Our numerical calculations also show that for a large variety of parameters (and in particular for the case of a Nb/Ti superlattice discussed in Sec. IV) the condition $Q_2 = 0$ cannot give higher T_c and H_{c2} than the condition $Q_1 = 0$. In particular, for the case $d_I = 0$ we have:

$$Q_1 = \frac{q_S}{q_I}\frac{1}{\eta_S}\tan\frac{q_S d_S}{2} - \frac{q_N}{q_I}\frac{1}{\eta_N}\tanh\frac{q_N d_N}{2} , \tag{17a}$$

$$Q_2 = -\frac{q_I}{q_S}\eta_S \tan\frac{q_S d_S}{2} - \frac{q_I}{q_N}\eta_N \tanh\frac{q_N d_N}{2} , \tag{17b}$$

so that $Q_2 < 0$ for realistic parameters and the condition $Q_1 = 0$ gives Eq. (14) with $\eta = \eta_S / \eta_N$.

IV. COMPARISON BETWEEN THEORY AND EXPERIMENT

We have used Eq. (12) for the theoretical explanation of the experimental results for the critical temperature of a Nb/Ti superlattice obtained by Qian *et al.*[4] In Fig. 2 the experimental curve $T_c(\Lambda)$ is plotted (with $d_S = d_N$), as well as the theoretical curve, which is in good agreement with the experimental data. As can be seen, there are three characteristic regions. For very thick S and N layers the critical temperature tends to the critical temperature of bulk niobium $T_{cS} = 9.22$ K. For very thin S and N layers T_c tends to the critical temperature of the interphase $T_{cI} = 9.5$ K.[4] For 30 Å $< \Lambda <$ 180 Å T_c is approximately constant and equal to 4 K, which would be the critical temperature of the Nb/Ti superlattice in the thin-layer limit in the absence of the alloy interphase between the Nb and Ti layers.

It should be noted that the theoretical curve in Fig. 2 is obtained for $T_{cN} = 0.4$ K, $d_I = 4$ Å, $\xi_S = 30$ Å, $\xi_I = 35$ Å, $\xi_N = 178$ Å, $\eta_S = 1.3$, and $\eta_N = 0.75$, i.e., with coherence lengths which are much smaller than corresponding bulk values (for example, $\xi_S \approx 170$ Å). Taking into account that $D = v_F l / 3$ where v_F is the Fermi velocity and l the mean-free path, this would mean [see Eq. (9)] that the mean-free paths and/or Fermi velocities are much smaller than in the bulk materials. Biagi *et al.*[3] have come to a similar conclusion for the perpendicular upper critical field of a Nb/Cu superlattice.[8] One possible explanation would involve electron scattering at the thin layers boundaries, the mean-free paths being suppressed and highly anisotropic.

In Fig. 3 the theoretical curves $H_{c2}(T)$ are plotted for different values of Λ as well as $H_{c2,S}(T)$, $H_{c2,N}(T)$, and

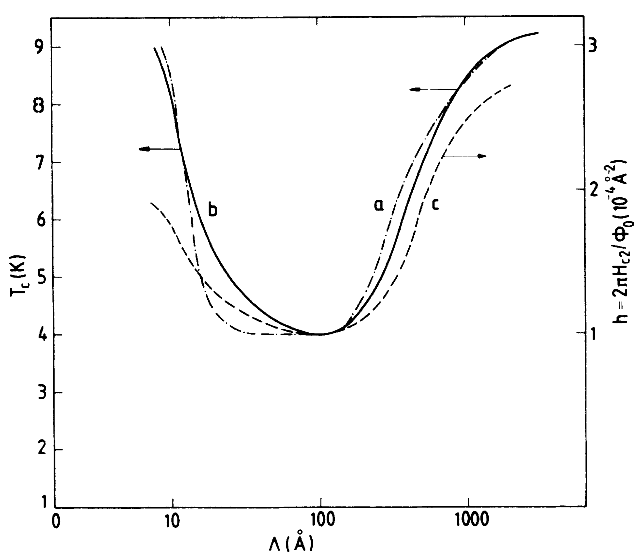

FIG. 2. Dependence of the critical temperature T_c and upper critical field $H_{c2}(T=0)$ upon the period $\Lambda = d_S + d_N + 2d_I$ for $d_S = d_N$ and constant d_I in a Nb/Ti superlattice: *a*, experimental results for T_c from Ref. 4 (dash-dot curve, left scale); *b*, theoretical result for T_c using parameters given in the text (solid curve, left scale); and *c*, theoretical prediction for $H_{c2}(T=0)$, expressed in reduced form as $h = 2\pi H_{c2}/\phi_0$, the square of the inverse magnetic length (dashed curve, right scale).

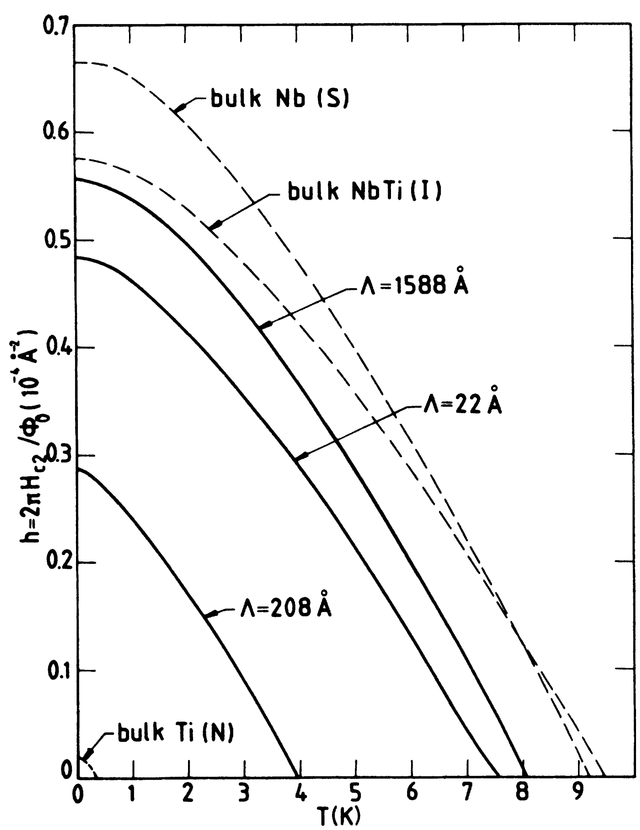

FIG. 3. Variation of the upper critical field H_{c2} (expressed in reduced form as $h = 2\pi H_{c2}/\phi_0$, the square of the inverse magnetic length) vs temperature T. Dashed curves show the intrinsic H_{c2} for bulk Nb (S), bulk Nb/Ti alloy (I) and bulk Ti (N). Solid curves show perpendicular H_{c2} calculated for the $S/I/N$ superlattice for parameters given in the text and $\Lambda = 1588$, 208, and 22 Å. Note that the curve for $\Lambda = 22$ Å is *above* the curve for $\Lambda = 208$ Å.

$H_{c2,I}(T)$. The parameters are the same as for the curve which represents $T_c(\Lambda)$ in Fig. 2. A significant positive curvature near T_c cannot be seen except for Λ corresponding to $T_c \gtrsim 8$ K, which is the temperature where $H_{c2,S}(T)$ and $H_{c2,I}(T)$ intersect. This effect has been discussed in Ref. 9.

The Λ dependence of H_{c2} can be seen from Fig. 2 where $H_{c2}(T=0)$ is plotted for the same parameters as for $T_c(\Lambda)$. This curve can be simply interpreted. For very large periods $H_{c2}(T=0)$ tends to $H_{c2,S}(T=0)$; if the periods are small, $H_{c2}(T=0)$ tends to $H_{c2,I}(T=0)$. The curve is significantly asymmetric because $H_{c2,I}(T=0)$ is much smaller than $H_{c2,S}(T=0)$, which is due to the fact that[3] $H_{c2,S,I} \sim \xi_{S,I}^{-2}$. Unfortunately, no experimental data for the perpendicular upper critical field of Nb/Ti superlattices are yet available for comparison with the theory. It would be especially interesting to see whether $H_{c2}(T=0)$ against Λ has "$T_c(\Lambda)$-like" profile which the theory predicts.

V. SUMMARY

We have extended the proximity-effect theory of a superlattice made of two different dirty metals, assuming that there is a thin alloy interphase between S and N layers. We have shown that the reduction of the superlattice problem to that of a trilayer seems to be correct as far as the critical temperature and the perpendicular upper critical field of a superlattice are concerned. We also have fitted our theory to the experimental data for a Nb/Ti superlattice, having taken much smaller mean-free paths than the bulk values. These discrepancies may be due to our neglect of anisotropy of l and, thereby, of the coherence lengths. The theory predicts a period dependence of the perpendicular upper critical field similar to that of the critical temperature, with a characteristic increase at small periods, where the interphase effect is significant.

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APPENDIX

In this appendix we derive the boundary-condition equation (12). The solutions of Eqs. (7b) and (7d) are [Fig. 1(b)]:

$$\begin{aligned}
&g_S(z)=A_S e^{iq_S z}+B_S e^{-iq_S z} \text{ in } S \text{ layers;}\\
&g_{I1}(z)=A_{I1} e^{iq_I z}+B_{I1} e^{-iq_I z} \text{ in "first" } I \text{ layers ;}\\
&(0<z<d_I,\ \Lambda<z<\Lambda+d_I,\ 2\Lambda<z<2\Lambda+d_I,\ \ldots);\\
&g_N(z)=A_N e^{q_N z}+B_N e^{-q_N z} \text{ in } N \text{ layers ;}
\end{aligned} \tag{A1}$$

and

$$g_{I2}(z)=A_{I2} e^{iq_I z}+B_{I2} e^{-iq_I z} \text{ in "second" } I \text{ layers}$$

$$(d_I+d_N<z<2d_I+d_N,\ \Lambda+d_I+d_N<z<\Lambda+2d_I+d_N,\ 2\Lambda+d_I+d_N<z<2\Lambda+2d_I+d_N,\ \ldots)\ ,$$

where the coefficients are different for different layers of the same metal.

Putting conditions (10) and (11) at I_2S, SI_1, I_1N, and NI_2 boundaries and taking into account that g must be a Bloch function

$$g(z)=e^{-iq_z\Lambda} g(z+\Lambda)\ , \tag{A2}$$

where $\Lambda=d_S+d_N+2d_I$, we obtain a system of eight linear homogeneous equations for eight unknown coefficients. After setting the determinant equal to zero, we get

$$\begin{aligned}
\cos(q_z\Lambda)=&\cos(q_S d_S)\cosh(q_N d_N)\cos(2q_I d_I)\\
&+\frac{1}{2}\left[\frac{q_I^2}{q_S q_N}\eta_S\eta_N-\frac{1}{\eta_S\eta_N}\frac{q_S q_N}{q_I^2}\right]\sinh(q_S d_S)\sinh(q_N d_N)\sin^2(q_I d_I)\\
&+\frac{1}{2}\left[\frac{q_N}{q_I}\frac{1}{\eta_N}-\frac{q_I}{q_N}\eta_N\right]\cos(q_S d_S)\sinh(q_N d_N)\sin(2q_I d_I)\\
&+\frac{1}{2}\left[\frac{q_N}{q_S}\frac{\eta_S}{\eta_N}-\frac{\eta_N}{\eta_S}\frac{q_S}{q_N}\right]\sin(q_S d_S)\sinh(q_N d_N)\cos^2(q_I d_I)\\
&-\frac{1}{2}\left[\frac{q_S}{q_I}\frac{1}{\eta_S}+\frac{q_I}{q_S}\eta_S\right]\sin(q_S d_S)\cosh(q_N d_N)\sin(2q_I d_I)\ .
\end{aligned} \tag{A3}$$

When $d_I=0$, (A3) reduces to the corresponding relation for bimetallic superlattices.[10,11] As in this case,[3,10] the lowest eigenvalue of Eqs. (7),

$$k_{S,I}^2-q_{S,I}^2=-k_N^2+q_N^2=2\pi H/\phi_0 ,$$

gives the largest field H_{c2} obtained from (A3) with $q_z=0$. In that case Eq. (A3) gives Eq. (12). Note also that if any q_i^2 ($i=S$, N, or I) is less than zero, we should simply replace q_i by iq_i in Eq. (12).

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