Boundary-condition effects on the superconducting transition temperature of proximity-effect systems

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The superconducting critical temperature, T_c, of different configurations of layers is studied under the de Gennes–Werthamer model. Certain inconsistencies are seen to develop with the use of this approach, calling into question previous results obtained. Preliminary results on Nb/Zr layered structures are also presented.

INTRODUCTION

The study of superconducting metallic multilayers has provided information on the dependence of the transition temperature on the period, Λ, of the multilayer.[1,2] Typically, the equations of de Gennes[3] and Werthamer[4] are used to understand the behavior of the results. The bilayer system, or "single-period" multilayer, has been studied in great detail. To extend these studies to many-period systems, one typically assumes that the infinite multilayer is a good approximation, which is equivalent to a bilayer with one-half the original thicknesses. It has been noted that a finite multilayer can be made equivalent to an infinite multilayer if the initial and final layers of the finite multilayer are one-half the thickness of the original layers.

In this paper, the de Gennes–Werthamer equations used in studies of the transition temperature for multilayer systems are solved for the M-period multilayer, where $M=2$ and 3, and for the symmetric systems $(NS)_M N$ and $(SN)_M S$, where M is the number of repetitions of NS or SN. Here, S and N refer to layers with the higher and lower transition temperature, respectively. The behavior of these systems is examined as M increases from one.

PROXIMITY-EFFECT EQUATIONS

For a system of multiple layers in the dirty limit with different transition temperatures, the de Gennes–Werthamer theory[3,4] gives the proximity-effect transition temperature for the system (T_c) as a solution of the following set of simultaneous equations for the gap function in each layer i:

$$-\chi(-\xi_i^2\nabla^2)\Delta_i(\mathbf{r})=\ln\left[\frac{T_c}{T_c^i}\right]\Delta_i(\mathbf{r}) , \tag{1}$$

where $\chi(z)=\psi(1/2+z/2)-\psi(1/2)$, $\psi(x)$ is the digamma function, T_c^i is the T_c of layer i in isolation, Δ_i is the gap function in layer i, and ξ_i is given by

$$\xi_i=\left[\frac{\hbar v_F^i l_i}{6\pi k_B T_c}\right]^{1/2} , \tag{2}$$

where v_F^i and l_i are the Fermi velocity and elastic mean free path in layer i. Using the relation[5] $v_F l=(\pi k_B/e)^2(\rho\gamma)^{-1}$, Eq. (2) becomes

$$\xi_i=\left[\frac{\pi\hbar k_B}{6e^2T_c}\frac{1}{\rho_i\gamma_i}\right]^{1/2} , \tag{3}$$

where ρ_i and γ_i are the low-temperature resistivity and electronic-specific-heat coefficient for layer i, respectively.

The gap function must also satisfy certain boundary conditions. For the de Gennes–Werthamer equations, at a metal-vacuum or metal-insulator interface the gap function obeys

$$\left.\frac{d\Delta_i}{dn}\right|_{\text{boundary}}=0 . \tag{4}$$

At a metal-metal interface in the dirty limit, de Gennes[3] has pointed out that the true boundary condition on the gap function is continuity of both Δ_i/N_iV_i and $(D_i/V_i)(d\Delta_i/dn)$, where N_i, V_i, and D_i are the density of states, pairing potential, and diffusion constant for the layer i. Since we are only interested in the transition temperature, we can use Werthamer's[4] combined version of the above boundary conditions:

$$\left.\frac{N_i\xi_i^2}{\Delta_i}\frac{d\Delta_i}{dn}\propto\frac{1}{\rho_i\Delta_i}\frac{d\Delta_i}{dn}\right|_{\text{continuous}} . \tag{5}$$

The only information lost in using this boundary condition is the magnitude of the gap function. For the case considered here, there are layers of two distinct materials, S and N, such that the proximity T_c obeys $T_c^N\le T_c\le T_c^S$. In this case, the gap function has the following form for the two layers:

$$\begin{aligned}&\Delta_i(x)\propto e^{iq_ix}+a_ie^{-iq_ix}, \quad \text{for } S ,\\ &\Delta_i(x)\propto e^{k_ix}+b_ie^{-k_ix}, \quad \text{for } N .\end{aligned} \tag{6}$$

Substituting these expressions into Eq. (1) gives the two fundamental equations for the decay lengths, q and k:

$$\chi(\xi_S^2q^2)=\ln\left[\frac{T_c^S}{T_c}\right], \quad -\chi(-\xi_N^2k^2)=\ln\left[\frac{T_c}{T_c^N}\right] . \tag{7}$$

Notice that every S and N layer has the same value of q

or k, which is one of the principal features of this approach. This point will be discussed later. Use of the above equations with those in Eqs. (4)–(6) allows one to evaluate T_c for the system.

For numerical computation in the example cases, the simplifying assumptions made in Werthamer's[4] work for the χ function are used,

$$\chi(z)\rightarrow\ln[1+(\pi^2 z/4)], \quad \text{for } z\geq 0$$
$$\rightarrow(\pi^2/4)\ln(1+z), \quad \text{for } z\leq 0 . \tag{8}$$

In addition the normal-state parameters for the two layers are assumed the same, with only $T_c^{S,N}$ varying, and are set equal to T_c^0 and 0, respectively. This gives for $\xi_{S,N}$

$$\xi_S=\xi_N=\xi_0\left[\frac{T_c^0}{T_c}\right]^{1/2}, \quad \xi_0=\left[\frac{\pi\hbar k_B}{6e^2T_c^0}\frac{1}{\rho\gamma}\right]^{1/2} . \tag{9}$$

For the calculations, distances will be expressed in terms of ξ_0, which is the "coherence" length for the S layers. The expressions for q and k, after substituting Eq. (8) into Eq. (7), become

$$q=\frac{2}{\pi\xi_0}[1-(T_c/T_c^0)]^{1/2} ,$$
$$k=\frac{1}{\xi_0}\left[\frac{T_c}{T_c^0}\right]^{1/2} \tag{10}$$

for $0\leq T_c\leq T_c^0$.

SOLUTIONS FOR M PERIODS ($M=1$, 2, AND 3)

In this section, the system considered is shown in Fig. 1(a), an M-period multilayer, with layer thicknesses d_S and d_N, with $\Lambda=d_S+d_N$. The system has a vacuum-metal interface at each end. The layer thicknesses are fixed, and only M varies. For the case of $M=1$, a bilayer, the solution obtained by substituting Eq. (6) into Eqs. (4) and (5) gives the well-known result

$$\frac{q}{\rho_S}\tan(q\,d_S)=\frac{k}{\rho_N}\tanh(kd_N), \quad \text{for } M=1 . \tag{11}$$

As M increases, one might expect that the system will begin to resemble an infinite multilayer. For an infinite periodic multilayer, it is known that the solution is the same as that for a regular bilayer with one-half the layer thicknesses, since the "vacuum-metal" boundary condition is met at the midpoint of the layers. For this case, the equation relating q and k is

$$\frac{q}{\rho_S}\tan\left[\frac{q\,d_S}{2}\right]=\frac{k}{\rho_N}\tanh\left[\frac{k\,d_N}{2}\right] ,$$
$$\text{for an infinite multilayer} . \tag{12}$$

Therefore, as M increases, the solutions for $T_c(M)$ should progress from the bilayer value down to the infinite multilayer value as shown in Fig. 2. The results of Fig. 2 are calculated using Eqs. (10)–(12), assuming $\rho_S=\rho_N$ and $d_S=d_N$.

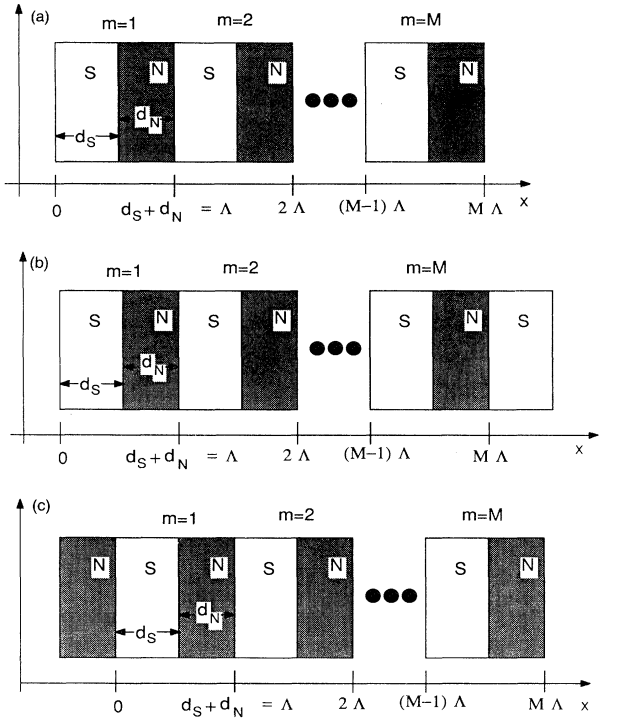

FIG. 1. (a) Schematic of an M-period multilayer $(NS)_M$ composed of layer S with thickness d_S, and layer N with thickness d_N. (b) Schematic of a $(SN)_MS$ system and (c) schematic of a $(NS)_MN$ system.

This progression can be studied starting from the $M=1$ point by applying the above equations to systems with more than one period. For simplicity, the definitions $q^*=q/\rho_S$ and $k^*=k/\rho_N$ are used. For the $M=2$ case, there are two exterior boundary conditions, and three interface boundary conditions. There are four unknown coefficients for the a's and b's from Eq. (6), and five equations. This leaves one equation relating q and k,

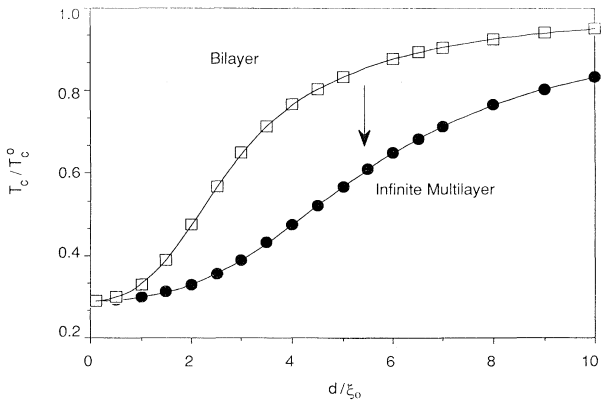

FIG. 2. Normalized T_c vs d/ξ_0 for the case of a bilayer and infinite multilayer. The lines are guides to the eye.

$$q^*[q^*\tan(q\,d_S)-k^*\tanh(k\,d_N)][k^*-q^*\tan(q\,d_S)\tanh(k\,d_N)]$$
$$=k^*[k^*\tanh(k\,d_N)-q^*\tan(q\,d_S)][q^*+k^*\tanh(kd_N)\tan(q\,d_S)]\ . \quad (13)$$

Obviously, the bilayer solution, Eq. (11), is a trivial solution to the above, giving $0=0$. Removing this solution from Eq. (13) gives

$$q^*[q^*\tan(q\,d_S)\tanh(k\,d_N)-k^*]$$
$$=k^*[q^*+k^*\tanh(k\,d_N)\tan(qd_S)]\ . \quad (14)$$

This equation can be solved as Eqs. (11) and (12) were. The results show that no solution exists for equal thickness systems with $d/\xi_0<1.70$, and for $d/\xi_0>3.0$, the solutions have the problem that $q^*d_S>\pi/2$, i.e., the gap function is oscillatory, which is unphysical. In between these two limits, the calculated T_c's are below those for the infinite multilayer, which is also unphysical. The only other solution to the $M=2$ equation was removed earlier, the bilayer solution, which is a trivial solution. In Fig. 3 the spatial dependence of the order parameter, $\Delta(x)/V(x)$, is schematically shown for the $M=1$ and $M=2$ cases when Eq. (11) is satisfied.

The same calculation has been carried out for the $M=3$ system, and the exact same solution occurs. I will not list the full expression relating q and k here. The point is that for $M>1$, the solution of the de Gennes–Werthamer equations, assumed valid for the $M=1$ case, lead to the $M=1$ solution plus a set of unphysical solutions. A similar result was seen in the work by Menon and Arnold.[6] Now the question arises as to whether the $M=1$ solution can be valid for $M>1$? The initial response, upon looking at the gap function, is no, since that picture of the gap function does not appear to make any physical sense. Namely, as one increases the size of the system, the resulting boundary conditions would give an exponentially increasing average of the order parameter. This would seem to violate the idea (from thermodynamics) that the order parameter should depend linearly on the system size, not exponentially, and (from Ginzburg-Landau theory) that the order parameter must remain small near the transition temperature. (The true behavior may depend upon a solution of the problem using the nonlinear terms in the proximity-effect equations.) Also, if Eq. (11) is the true solution for all M, it would imply that the boundary conditions on the problem have a dramatic effect for the case of large M. It would seem at this point that there is some type of inconsistency. This problem can be looked at in another light, however, in the hopes of seeing what the true behavior for $M>1$ will be.

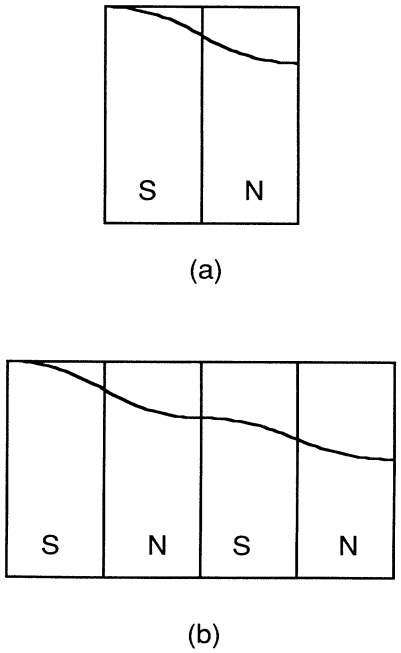

FIG. 3. Schematics of the order parameter $\Delta(x)/V(x)$, for (a) a bilayer and (b) a 2-period multilayer. Here, $N(x)$ is assumed constant throughout.

SOLUTIONS FOR THE SYMMETRIC SYSTEMS: *NS . . . N* AND *SN . . . S*

For the $(NS)_M$ case, there is no obvious point of symmetry, which exists for the two cases of $N(SN)_M$ and $S(NS)_M$ systems, as shown in Figs. 1(b) and 1(c). The problem can be simplified considerably, and studied with the same analysis as was done earlier. What one finds rather quickly is that there is a simple relation between q and k for any value of M. First consider the $S(NS)_M$ system. The solution for the $M=1$ case, is simply

$$\frac{k^*}{q^*}\tanh\left[\frac{k\,d_N}{2}\right]=\tan(q\,d_S),\quad \beta_1=0\ , \quad (15)$$

where the need for β_1 will be explained later. The solution for the $M=2$ case now involves solving the boundary conditions at two interfaces, and the solution is

$$\beta_2=\frac{q^*}{k^*}\tan(q\,d_S)\ ,$$
$$\frac{q^*}{k^*}\tan\left[\frac{q\,d_S}{2}\right]=\frac{\tanh(k\,d_N)-\beta_2}{1-\beta_2\tanh(k\,d_N)}\ . \quad (16)$$

Continuing in this vein, one finds that for general m, the solution can be arrived at by solving the following: For M even,

$$\beta_{M\ \text{even}}=\frac{q^*}{k^*}\left[\frac{\tan(q\,d_S)-\beta_{M-1}}{1+\beta_{M-1}\tan(q\,d_S)}\right]\ ,$$
$$\frac{q^*}{k^*}\tan\left[\frac{q\,d_S}{2}\right]=\frac{\tanh(k\,d_N)-\beta_M}{1-\beta_M\tanh(k\,d_N)}\ . \quad (17a)$$

For M odd,

$$\beta_{M\ \mathrm{odd}}=\frac{k^*}{q^*}\left[\frac{\tanh(k\,d_N)-\beta_{M-1}}{1-\beta_{M-1}\tanh(k\,d_N)}\right],$$
$$\frac{k^*}{q^*}\tanh\left[\frac{k\,d_N}{2}\right]=\frac{\tan(q\,d_S)-\beta_M}{1+\beta_M\tan(q\,d_S)}. \tag{17b}$$

The solutions for the $N(SN)_M$ system are derived in the exact same manner, and are quite similar. The general solution is (with $\alpha_1=0$, as before) for M even,

$$\alpha_{M\ \mathrm{even}}=\frac{k^*}{q^*}\left[\frac{\alpha_{M-1}+\tanh(k\,d_N)}{\alpha_{M-1}\tanh(k\,d_N)+1}\right],$$
$$\frac{k^*}{q^*}\tanh\left[\frac{k\,d_N}{2}\right]=\frac{\tan(q\,d_S)-\alpha_M}{1+\alpha_M\tan(q\,d_S)}, \tag{18a}$$

and for M odd,

$$\alpha_{M\ \mathrm{odd}}=\frac{q^*}{k^*}\left[\frac{\alpha_{M-1}-\tan(q\,d_S)}{1+\alpha_{M-1}\tan(q\,d_S)}\right],$$
$$\frac{q^*}{k^*}\tan\left[\frac{q\,d_S}{2}\right]=\frac{\alpha_M+\tanh(k\,d_N)}{\alpha_M\tanh(k\,d_N)+1}. \tag{18b}$$

With the above set of equations, the progression of the proximity-effect T_c as M increases can be studied for both the $(NS)_MN$ and $(SN)_MS$ systems. Again, the same simplifications are made as before, and the case of $d_S=d_N=d$ is considered. Figure 4 shows the results of the calculations for both sets of systems with $M=1$, 2, 3, and 4, and compares them to the results for the bilayer and the infinite multilayer for $\Lambda=2d$ varying from 0 to $10\xi_0$. What one immediately sees, is that instead of these two systems approaching the same value of T_c as M increases, they each tend towards a different limit. The $(NS)_MN$ system tends towards the infinite multilayer result, while the $(SN)_MS$ system tends towards the bilayer result. Figure 5 shows this more clearly by looking at the two cases for fixed values of d as M varies.

The conclusion from these results is that the existence of a center of symmetry does not remove the inconsistency from these equations. For values of M used in multilayer systems, the calculations show that the transition temperatures of the two systems do not tend together, leaving the behavior of T_c as a function of the number of periods for a multilayer unresolved.

The use of a single value of q or k for all the S or N layers in these systems, known as the principal root approximation,[7] would seem to be a major cause of these inconsistencies. From Fig. 3(b), one would expect that the curvature in the order parameter would be different for the two S or two N layers, since one of the layers has to meet an external boundary condition. However, the usual de Gennes–Werthamer approach cannot incorporate this, especially since the transition temperature determines the curvature in the layers, and that is assumed the same throughout. The same problem arises in the symmetric systems, $(SN)_MS$ and $(NS)_MN$, for $M>1$. Perhaps a variational approach similar to that in Ref. 4 may indicate the behavior of multiple layer systems.

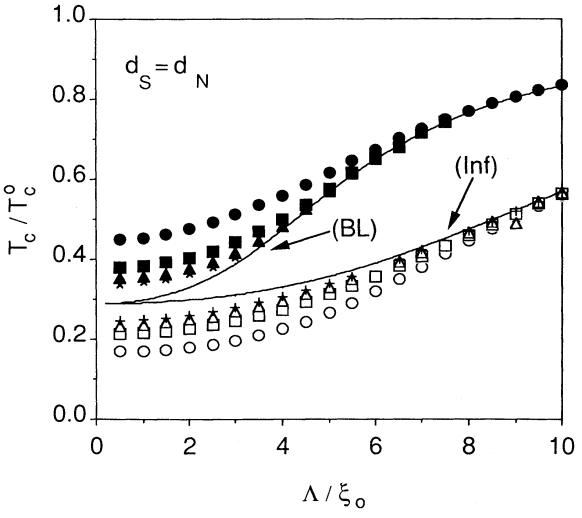

FIG. 4. Normalized T_c vs Λ/ξ_0 for the case of $(SN)_MS$ and $(NS)_MN$ systems with equal thickness layers for $M=1$–4 compared to the bilayer (BL) and infinite multilayer (Inf) results. The solid symbols and x refer to the $(SN)_MS$ system, and the open symbols and + refer to the $(NS)_MN$ system; with $M=1$, 2, 3, an 4 represented by circles, squares, triangles, and crosses, respectively.

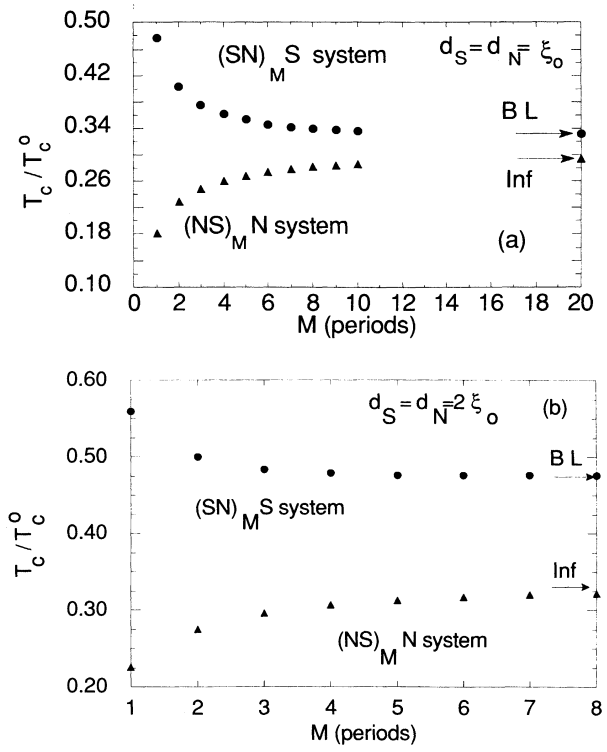

FIG. 5. Normalized T_c for the $(SN)_MS$ and $(NS)_MN$ systems vs the number of periods for (a) $d_S=d_N=\xi_0$ and (b) $d_S=d_N=2\xi_0$. The arrows indicate the calculated values for the bilayer (BL) and infinite multilayer (Inf) case.

TABLE I. T_c as a function of number of periods: $\Lambda = 1200$ Å; $d_{Nb} \approx d_{Zr}$; T_c (SN bilayer) = 8.74 K; and T_c (infinite multilayer) = 8.22 K.

Number of periods	T_c (K) measured
1	8.75
2	8.77
3	8.81

TABLE II. Parameters used in bilayer fit to Nb/Zr.

	Nb	Zr
T_c (K)	9.2	0.8
γ^* (ergs/cm^3 K^2)	7200	1856
ρ ($\mu\Omega$ cm)	3.2	25

PRELIMINARY EXPERIMENTAL RESULTS

To study the above calculations, three multilayers of Nb/Zr were made and measured. The fabrication of the samples is identical to that in earlier work.[8,9] Nb/Zr was used for two reasons: first, it lies well in the dirty limit, and there are no problems with clean effects, and second, the predicted difference between the bilayer T_c and the infinite multilayer T_c is large. The three samples had $d_{Nb} \approx d_{Zr}$ and $\Lambda = 1200$ Å, and were made with 1, 2, and 3 periods. A 550-Å-thick film of Nb deposited under identical conditions had at T_c of 8.89 K and a residual resistivity ratio of 9.45. The results for these samples are given in Table I along with the calculated T_c's from the bilayer and infinite multilayer models. For this case, the full equations for $\chi(z)$ were used, and the parameters used to calculate the T_c's were taken from transport measurements made on these and similar samples and from specific-heat measurements (Refs. 8 and 9) and are listed in Table II. Notice we have used the full T_c of niobium for the calculations, instead of the T_c of the thin Nb film, for comparison to the earlier work. Good agreement with the bilayer value for T_c is seen. Notice that as M increases, T_c actually goes up, not down. The latter rise is very slight, and may be due to sample variation. What is completely clear is that there is no decrease towards the typical infinite multilayer result, and that experimentally, it seems that the T_c for the $M > 1$ case may be the same as the $M = 1$ case, even if the picture of the order parameter is unphysical.

CONCLUSIONS

In this paper, the standard de Gennes–Werthamer approach to calculate the transition temperature of multiple layer systems, of either the type $(NS)_M$, $(SN)_M S$, or $(NS)_M N$ has been carried out. For the $(NS)_M$ systems, it is found that the solution is the same for all values of M as it is for $M = 1$, leading to an unphysical picture of the order parameter. The $(SN)_M S$ and $(NS)_M N$ do not have this problem, but as M increases, the two systems do not approach each other, and instead the $(SN)_M S$ system approaches the standard bilayer result, while the $(NS)_M N$ system approaches the "traditional" infinite multilayer result. The use of the principal root approximation is seen as a possible cause of the difficulty. Experimentally, the progression of T_c was observed in Nb/Zr structures of the type $(NS)_M$ with $M = 1$, 2, and 3. T_c was found to remain nearly constant, agreeing with the calculated results. Obviously, more experimental and theoretical work remain to be done on this problem.

ACKNOWLEDGMENTS

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