Localized surface moments and a magnetic phase transition: The source of the anomalous H_{c3}/H_{c2} temperature dependence for clean strain-free vanadium specimens

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It is suggested that localized surface moments are the source of the anomalous sheath-state superconductivity exhibited by clean, well annealed vanadium specimens in H_{c3}/H_{c2} vs T data. I propose a simple, heuristic combination of the normal surface-layer modification of the sheath state, as calculated by de Gennes, and Werthamer's theory of the proximity effect of magnetic impurities on superconducting films. A magnetic phase transition of localized surface moments is also assumed. This results in an expression for a temperature-dependent extrapolation length $b(T)$ which shows good agreement with published values extracted from "linearized" H_{c3}/H_{c2} data. The values of the parameters of the model used in the fit are consistent with (1) the low-temperature susceptibility data of Akoh and Tasaki for vanadium micropowders, (2) the results of the microscopic calculation of fluctuation-induced surface moments by Grempel and Ying, and (3) the estimate for the diffusion length of Cooper pairs into a magnetic layer coating a clean superconducting surface by Hauser, Theurer, and Werthamer.

I. INTRODUCTION

Akoh and Tasaki[1] have reported experimental evidence of magnetic moments localized in the free surface (1–2 atomic layers) of vanadium micropowder specimens. Subsequently, Grempel and Ying[2] have performed a spin-fluctuation calculation for the electronic surface states of vanadium. Their results predict fluctuation-induced, localized surface moments which obey Curie's law. An antiferromagnetic exchange-interaction strength was also calculated, indicative of a magnetic transition temperature of 3.8 K. It is my conjecture that the experimental results and theoretical interpretation described above also allow for a more physical explanation than has heretofore been advanced for the finite value of the extrapolation length b extracted from sheath-state H_{c3}/H_{c2} data, reported for clean, freshly etched, vanadium bulk specimens.[3,4]

The extrapolation length b is a measure of mixed boundary conditions at a superconducting interface: $b \equiv \Psi_0/(d\Psi/dx)_0$, where Ψ_0 and $(d\Psi/dx)_0$ are, respectively, the Ginsburg-Landau (GL) order parameter and its normal derivative at the surface of the superconductor.[5] Finite values for b are required in the linear GL theory for $H_{c3}/H_{c2} \neq 1.69$ as $T \rightarrow T_c$. It possibly needs to be reiterated that the H_{c3}/H_{c2} data of Ref. 4 are "linearized" according to the prescription

$$\left(\frac{H_{c3}}{H_{c2}}\right)_l = \left(\frac{H_{c3}}{H_{c2}}\right)_e \times \frac{1}{C(T)} ,$$

where T is the temperature, $(H_{c3}/H_{c2})_e$ the experimental value of the critical field ratios, $C(T)$ a polynomial in T that is the result of a microscopic calculation by Hu and Koreman[6] which effectively gives the nonlinear Ginsberg-Landau contributions to the H_{c3}/H_{c2} ratio, and $(H_{c3}/H_{c2})_l$ the resultant "linearized" values of H_{c3}/H_{c2}. The success of the procedure is evidenced by the excellent fit of a calculation of H_{c3}/H_{c2} using the linear GL theory weighted by $C(T)$ to the data of Ref. 4 for $T \geq 3$ K.

The theoretical treatment of choice that has previously been employed to explain $H_{c3}/H_{c2} \neq 1.69$ near T_c for clean, homogeneous specimens is one proposed by Hu.[7] In this model b is expressed in terms of the enhancement or depression of the electron-electron interaction strength ΔV over a surface region of depth d. Specific sources of the modified electronic states of the surface region have thus far not been identified, especially for the pure, well annealed specimens of Nb and V to which Hu's phenomenology has been applied.[3,4] Furthermore, the suggestion has been previously advanced in Ref. 4 that the abrupt departure of the good fit at $T \cong 3$ K may be interpreted in the context of the Hu model as evidence of a very deep "A" region; i.e., the depth of the perturbed surface region d is of the order of ξ, the superconducting coherence length at that temperature 675 Å. This leaves one with the puzzle of explaining an intrinsically modified surface region of such great depth.

I now suggest that the H_{c3}/H_{c2} data near T_c of Ref. 4 can be understood in the context of a model in which the pair-breaking perturbation is localized spins at the free surface of the vanadium ($d = 3$–6 Å). It is further suggested that the abrupt decrease in b at $T \cong 3$ K required to fit the data is associated with the onset of magnetic ordering of the surface moments and a concomitant increase in their pair-breaking strength. Such ordering was inferred by Akoh and Tasaki[1] to occur at $T < 4$ K for their vanadium micropowder specimens; and, as already indicated, Grempel and Ying[2] estimated an antiferromagnetic transition temperature of $T \cong 3.8$ K in their fluctuation-induced surface moment calculation. Thus, the thrust of this paper is to provide a plausible explanation for the finite value of the extrapolation length b and its temperature dependence numerically calculated from the integral form of the linear GL equations with $(H_{c3}/H_{c2})_l$ as the input parameter.

II. MODEL

The simple model that I wish to present is essentially a synthesis of two related calculations. The first is that of de

Gennes and Sarma for the extrapolation length b of a superconducting surface coated with a normal metal.[5] The second is Werthamer's calculation of the proximity effect of localized moments on the surface of a superconducting film, i.e., the reduction of the transition temperature of the film.[8]

de Gennes' expression for b is given in terms of the normal layer–superconducting substrate electronic parameters:

$$\frac{1}{b}=\frac{\sigma_n}{\sigma_s}K\tanh(d_nK) \ , \tag{1}$$

where σ_n and σ_s are the electrical conductivity of the normal and superconducting materials, respectively, d_n the thickness of the normal layer, and K^{-1} the diffusion length of the Cooper pairs into the normal region. In this model I assume that the "normal" region consists *only* of fluctuation-induced, localized moments that extend to a depth of 1–2 atomic layers—in effect, a clean strain-free surface.

I next assume that Werthamer's proximity effect calculation may be applied in order to develop a simple relationship between the diffusion length K^{-1} and the mean free path l_n of the "normal" layer.[8,9] For the "normal" surface layer assumed, Eq. (9) of Ref. 8 reduces to

$$K^{-1}=(D_n\tau_n)^{1/2}=\frac{v_F\tau_n}{\sqrt{3}}=\frac{l_n}{\sqrt{3}} \ , \tag{2}$$

where D_n is the "normal" region diffusion coefficient, $1/\tau_n$ the rate of pair destruction by the localized moments via the exchange interaction, and v_F is the Fermi velocity. The resulting expression[10] for the extrapolation length b from the combination of Eqs. (1) and (2), and from $\sigma_n/\sigma_s=(N_n/N_s)^2 l_n/l_s$, is

$$\frac{1}{b}=\frac{\sqrt{3}}{l_s}\left(\frac{N_n}{N_s}\right)^2\tanh\left(\frac{\sqrt{3}d_n}{l_n}\right) \ , \tag{3}$$

where now l_s and l_n are, respectively, the interior and surface electronic mean free paths and N_n/N_s the ratio of the surface and interior density of states, a quantity calculated by Grempel and Ying.[2]

I now assume an additional pair breaking mechanism associated with the onset of ordering of the localized moments at a temperature T_m and, further, that the strength of the "ordered" pair-breaking component is proportional to the magnetic order parameter $m(t)$. Therefore, the effective pair-breaking rate $1/\tau_n'$, for $T<T_m$ is now given by

$$\frac{1}{\tau_n'}=\frac{1}{\tau_n}+\frac{m(t)}{\tau_0} \ , \tag{4}$$

where $m(t)/\tau_0$ is the increment in the rate of pair breaking associated with magnetic ordering and t is the reduced magnetic temperature (T/T_m). For reasons of clarity and ease of calculation, I take $m(t)$ to be the magnitude of the normalized, mean-field theory magnetization for spin-$\frac{1}{2}$ moments: $m(t)=\tanh[m(t)/t]$. Finally,

$$\frac{1}{b(t)}=\frac{\sqrt{3}}{l_s}\left(\frac{N_n}{N_s}\right)^2\tanh\left[\frac{\sqrt{3}d_n}{l_n}\left(1+\frac{l_n m(t)}{l_0}\right)\right] \ , \tag{5}$$

where $l_0\equiv v_F\tau_0$ and may be interpreted as the mean free path associated with magnetic ordering in the "normal" surface layer.

TABLE I. Values of parameters used in fit of Eq. (5) to ξ_0/b data.

$l_s=9030$ Å	(See Ref. 4)	$d_n/l_n=0.31$
$\xi_0=450$ Å	(See Ref. 4)	$l_n/l_0=0.52$
$N_s/N_n\cong 0.7$	(See Ref. 2)	$T_m=3$ K
$d_n=3$–6 Å	(See Refs. 1 and 2)	

III. RESULTS AND DISCUSSION

The adjustable parameters of this model are the ratios of lengths characteristic of the "normal" surface region, d_n/l_n and l_n/l_0, and the magnetic transition temperature T_m of this region. Values of these parameters as determined by a least-square fit of Eq. (5) to the published values of ξ_0/b are also given in Table I. [The extrapolation length calculations of Ref. 4 were checked resulting in slight corrections ($<5\%$) to the previously published values.] The resulting reasonably good fit is shown in Fig. 1. The value chosen for the magnetic transition temperature $T_m=3$ K is comparable to the approximate calculation by Grempel and Ying[2] of a Néel temperature of 3.8 K and consistent with the magnetic ordering temperature that may be inferred from Akoh and Tasaki's[1] data. $K^{-1}\cong 5.7$–11.4 Å, is calculated from the assumed value for $d_n=3$–6 Å chosen to agree with the results of Ref. 1 and the calculation of Ref. 2, the fitted value of the ratio d_n/l_n, and Eq. (2). This value is consistent with the calculation of Hauser and co-workers of K^{-1} from proximity effect measurements on magnetic material-superconducting film sandwiches.[8] For an Fe-Pb film it was estimated that $K^{-1}\cong 6$ Å. On the other hand, the increased degradation of the sheath-state superconductivity that accompanies the onset of magnetic ordering, as indicated by the fitted value of $l_n/l_0\cong 0.5$, is not so easily understood. Both the measurements[1] and calculations[2] of localized moments on the free surface of vanadium are indicative of antiferromagnetic ordering and it has been argued that such ordering in the bulk should reduce the exchange scattering by localized spins of Cooper pairs.[5] Nevertheless,

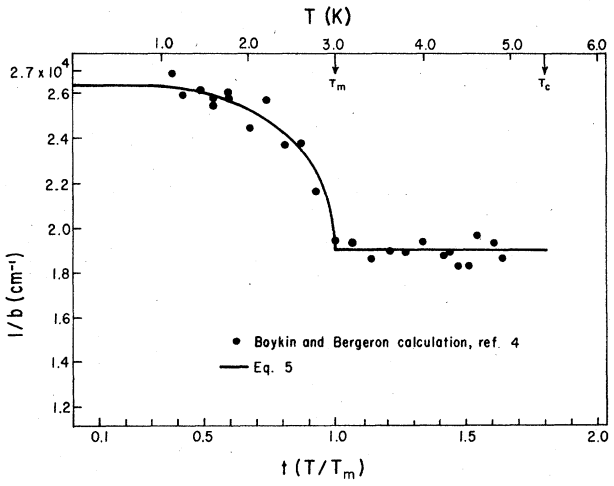

FIG. 1. Inverse extrapolation length vs temperature and reduced magnetic temperature. The solid line is a plot of Eq. (5) and the points are calculated values of $1/b$ from Ref. 4.

the increase in pair scattering associated with surface magnetic ordering that is implied by the fit of this model to $b(t)$ from H_{c3}/H_{c2} data is certainly consistent with the results of Hauser and co-workers for superconducting antiferromagnetic composite films which showed decreases in the transition temperature comparable to that achieved by ferromagnetic coatings or localized moments imbedded in a normal metal coating.[8]

The results of this calculation are further testable[10] through the linear dependence of the extrapolation length b on the interior or bulk mean free path l_s given by Eq. (5). The terms in the coefficient of l_s are relatively insensitive to the purity level of the specimen provided that $l_s \gg l_n$ or l_0 and that the free surface is clean and unstrained. Thus, measurements of H_{c3}/H_{c2} as a function of temperature on a series of vanadium specimens of varying residual resistance ratio R_R $(25 < R_R < 500)$ would allow for a test of the model. The only other published H_{c3}/H_{c2} data for vanadium of which I have knowledge are for an annealed, polycrystalline specimen whose $R_R = 720$ ($l_s = 1.3 \times 10^4$ Å).[3] The implied [through Eq. (5)] values for b of this specimen result in an H_{c3}/H_{c2} temperature dependence that is not experimentally distinguishable from the data analyzed here or presented in Ref. 3.

Finally, it is an interesting speculation that the observed temperature dependence of H_{c3}/H_{c2} in well annealed, freshly etched niobium specimens[3,11] may have a similar physical origin to that proposed herein for vanadium—localized, fluctuation-induced surface moments that depress sheath-state superconductivity and result in a finite value for the extrapolation length or the mixed boundary condition parameter b. This conjecture can of course be tested directly by a search for surface moments in niobium specimens with reasonably large surface to volume ratios, and indirectly by measurements of the temperature dependence of H_{c3}/H_{c2} for a series of niobium specimens of varying purity levels.

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[10]The first test of this model presents itself at this point since,

$$\frac{l_s}{\sqrt{3}b(t)}\left(\frac{N_s}{N_n}\right)^2 = \tanh\left(\frac{\sqrt{3}d_n}{l_n}\right) \leq 1 \ .$$

The data listed in Table I and the experimentally derived maximum value of $1/b(t)$ shown in Fig. 1 satisfy this constraint, with the left-hand side of the equality $= 0.68$, for $(1/b(t))_{\max} \cong 1/b(0$ K).

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