Theoretical model of superconductivity and the martensitic transformation in $A15$ compounds*

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(Received 8 April 1976)

The effect of the martensitic transition on superconductivity in the $A15$ compounds is examined theoretically. The BCS pairing potential is added to the Peierls potential in the Gorkov model of the martensitic transition. We find that the two potentials compete to open an energy gap on the common portion of Fermi surface and that one phase transition depresses the other. It is found that the $T_c = 17$ K in V_3Si includes a 0.3-K reduction due to this competition and that superconductivity arrests the development of tetragonality below 17 K. It is predicted that at high pressure (~ 25 kbar) the martensitic transition will be precluded.

I. INTRODUCTION

Intermetallic compounds of the $A15$ crystal structure exhibit the highest known superconducting transition temperatures.[1] Several of these high-T_c compounds undergo a structural "martensitic" transition at a higher temperature T_m.[2] In this paper, we examine the effect of the structural transition on superconductivity.

At the martensitic transition, the cubic $A15$ unit cell is distorted to a tetragonal cell with paired transition-metal atoms in two of the three linear chains.[3] This observation led Gorkov[4] to suggest that the martensitic transition is driven by a Peierls instability in the linear chains. In the cubic $A15$ structure the electronic energy bands are doubly degenerate at the X point (the center of the face of the simple-cubic Brillouin zone). This X-point degeneracy is split by the Peierls-like energy gap arising from a tetragonal distortion of the crystal. If the Fermi surface lies near the X point, the electronic energy is reduced by the Peierls gap and this energy, according to the Gorkov model, drives the structural phase transition. A Landau theory based on Gorkov's picture is in good agreement with a number of experiments on V_3Si and Nb_3Sn.[5] The Gorkov model for the martensitic transition in the $A15$ compounds is closely related to the charge-density-wave transitions observed in transition-metal dichalcogenides,[6] and to the Peierls transition found in tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ).[7]

As discussed briefly in a previous paper,[5] the structural transition affects the superconducting transition in two ways. The elevation of T_c by the softening of phonons has been understood qualitatively for a long time, though as yet there is no quantitative estimate of its importance. In an appreciable fraction of the Brillouin zone, phonons soften as the structural transition is approached, and recover after passing into the tetragonal phase.[3] The energy of the long-wavelength [110] phonon with transverse [1$\bar{1}$0] polarization almost vanishes at T_m. Since the coupling constant for superconductivity is proportional to electronic constants divided by the mean-square phonon energy,[8] any softening of the phonon spectrum enhances the coupling constant and raises T_c. Secondly, in the tetragonal phase, T_c is depressed by the presence of the Peierls gap. This is a new effect which follows from the Gorkov model. Near the X point, the Peierls gap separates the energy bands, depopulating that area of the Fermi surface and reducing the number of electrons available for BCS pairing. A substantial fraction of the electronic density of states at E_F can be removed by the Peierls gap, and this lowers T_c.

In this paper we present a microscopic theoretical model in which the BCS gap and the Peierls gap can coexist. The model is intended to be realistic enough to describe the $A15$ compounds, yet simple enough to allow easy solution. We argue that the details of the band structure are unimportant for the energetics of the phase transitions, and that one need only know the densities of states of the electrons participating in the transitions and the relevant coupling constants. These parameters can all be found from experiment. We then calculate the phase diagram of the crystal and the temperature dependence of the two gaps. We find that the two energy gaps compete for the same portion of the Fermi surface, and that each inhibits the other. For parameters appropriate to V_3Si ($T_m = 21$ K, $T_c = 17$ K), we find that at the superconducting transition the developing tetragonality is arrested and is subsequently depressed slightly. We further find that if the superconducting transition occurs in the cubic crystal (as it should in V_3Si at high pressure), the martensitic transition is precluded and the crystal always remains cubic.

With this model we can study only the effect of the Peierls gap on superconductivity. Since the

BCS interaction is taken as a phenomenological parameter, we can learn nothing about the effect of soft phonons.

II. MODEL HAMILTONIAN

We treat a model of electrons in Bloch states in the presence of a static lattice distortion and interacting via a BCS pairing interaction.[9] The Hamiltonian is the sum of several terms

$$\mathcal{H} = \mathcal{H}_E + \mathcal{H}_0 + \mathcal{H}_p + \mathcal{H}_s , \tag{1}$$

where $\mathcal{H}_0$ describes the conduction band in the normal cubic phase:

$$\mathcal{H}_0 = \sum_{kn\sigma} \epsilon_{kn} c^\dagger_{kn\sigma} c_{kn\sigma} , \tag{2}$$

where $c^\dagger_{kn\sigma}$ and $c_{kn\sigma}$ create and annihilate electrons in Bloch states with crystal momentum k, band index n, and spin σ. $\mathcal{H}_p$ is the electron-phonon coupling which precipitates the Peierls transition

$$\mathcal{H}_p = -\sum_{k\sigma nn'} W_{knn'} c^\dagger_{kn\sigma} c_{kn'\sigma} , \tag{3}$$

where $W_{knn'}$ is a matrix element proportional to the lattice distortion (acoustic plus optical phonon). $\mathcal{H}_p$ is important only near the X point, where it breaks the degeneracy between bands (labeled + and −). Here we can approximate $W_{k+-} = W_{k-+} = W$ independent of k. W will be referred to as the Peierls gap, though the actual separation between bands at the X point is $2W$ (in the normal phase). The crystal opposes the distortion with an elastic energy proportional to the square of the distortion, or to W^2. This energy is contained in

$$\mathcal{H}_E = (N/V_p) W^2 , \tag{4}$$

where N is the total number of atoms in the crystal and V_p is proportional to the relevant electron-phonon matrix element squared, divided by the relevant elastic constant. Finally, $\mathcal{H}_s$ is the familiar phonon-mediated electron-electron interaction responsible for superconductivity:

$$\mathcal{H}_s = -\sum_{kk'nn'} V_{kk'nn'} c^\dagger_{k'n'\uparrow} c^\dagger_{-k'n'\downarrow} c_{-kn\downarrow} c_{kn\uparrow} . \tag{5}$$

We use the BCS model interaction which approximates $V_{kk'nn'}$ as a constant, $V_{\rm BCS}/N$, for ϵ_{kn} and $\epsilon_{k'n'}$ within some $\hbar\omega_0$ of the Fermi energy, and as zero, otherwise.

The band structure along the Γ-X line near the (100) X point is

$$\epsilon_{k\pm} = \pm \hbar v_F \left| \hat{x} \cdot \vec{k} - \pi/a \right| , \tag{6}$$

where a is the cubic lattice constant and the X-point energy is taken to be the zero of energy. Equation (6) is a consequence of crystal symmetry and is an exact limit. The Peierls gap affects electrons within kT of the Fermi energy and only in a small sector of momentum space near the X point. This region we name region 1, the rest of the Fermi surface is contained in region 2. As a simplification we assume that Eq. (6) obtains in region 1, and that in region 2 the energy bands are independently linear in k. We also assume that in region 2 the W matrix vanishes. Finally, we assume that the Fermi energy coincides with the X-point energy so that the model has electron-hole symmetry. This last approximation forces the martensitic transition to be second order instead of weakly first order,[5] and the other approximations probably introduce further small errors into the temperature dependence of the energy gaps. We believe that, except for minor details, the theory realistically models both phase transitions in the $A15$ compounds.

III. TRANSFORMATION TO QUASIPARTICLE COORDINATES

In region 2 of momentum space, where the Peierls gap is unimportant, electrons of momentum p and spin up are mixed with holes of momentum $-p$ and spin down by the BCS pairing term in the Hamiltonian. The Hamiltonian is brought into diagonal form by the Bogoliubov transformation[10] to quasiparticle coordinates.

In region 1 the situation is complicated by the additional presence of the Peierls gap. The commutators are

$$\begin{aligned}
[\mathcal{H}, c_{p-\uparrow}] &= -\epsilon_{p-} c_{p-\uparrow} + W c_{p+\uparrow} + \Delta_p c^\dagger_{-p-\downarrow} , \\
[\mathcal{H}, c^\dagger_{-p-\downarrow}] &= \epsilon_{p-} c^\dagger_{-p-\downarrow} - W c^\dagger_{-p+\uparrow} + \Delta_p c_{p-\uparrow} , \\
[\mathcal{H}, c_{p+\uparrow}] &= \epsilon_{p-} c_{p+\uparrow} + W c_{p-\uparrow} + \Delta_p c^\dagger_{-p+\downarrow} , \\
[\mathcal{H}, c^\dagger_{-p+\downarrow}] &= -\epsilon_{p-} c_{-p+\downarrow} - W c^\dagger_{-p-\downarrow} + \Delta_p c_{p+\uparrow} ,
\end{aligned} \tag{7}$$

which shows that four excitations are mixed by the Hamiltonian: electrons at p with spin up in the + and − bands, and holes at $-p$ and with spin down in the + and − bands. The BCS interaction mixes electrons and holes, and the Peierls term mixes the + and − bands. The normal modes are obtained from a generalized Bogoliubov transformation. We define quasiparticle operators A_p, B_p, C_p, and D_p by the relations

$$\begin{pmatrix} A_p \\ B^\dagger_p \\ C^\dagger_p \\ D_p \end{pmatrix} = \begin{pmatrix} u_p & -v_p & f_p & -g_p \\ v_p & u_p & g_p & f_p \\ -f_p & -g_p & u_p & v_p \\ g_p & -f_p & -v_p & u_p \end{pmatrix} \begin{pmatrix} c_{p-\uparrow} \\ c^\dagger_{-p-\downarrow} \\ c_{p+\uparrow} \\ c^\dagger_{-p+\downarrow} \end{pmatrix} . \tag{8}$$

The coefficients are taken real and they satisfy

$$u_p^2+v_p^2+f_p^2+g_p^2=1\,, \tag{9}$$

ensuring that the quasiparticle operators obey fermion anticommutation rules. The matrix is orthogonal, so its inverse is its transpose. The condition that the quasiparticles be noninteracting excitations is

$$[\mathcal{H},A_p]+\Omega_p A_p=0\,; \tag{10}$$

the analogous equations for B_p, C_p, or D_p are equivalent to this and they are all satisfied provided

$$\begin{bmatrix} \Omega_p-\epsilon_{p-} & -\Delta_p & W & 0 \\ \Delta_p & -\Omega_p-\epsilon_{p-} & 0 & W \\ W & 0 & \Omega_p+\epsilon_{p-} & -\Delta_p \\ 0 & W & \Delta_p & -\Omega_p+\epsilon_{p-} \end{bmatrix} \begin{bmatrix} u_p \\ v_p \\ f_p \\ g_p \end{bmatrix} =0\,. \tag{11}$$

The condition for a solution is that the determinant vanish and that fixes the eigenvalue

$$\Omega_p=(\epsilon_{p-}^2+\Delta_p^2+W^2)^{1/2}\,. \tag{12}$$

The remaining twofold degeneracy permits the additional constraint $v_p/u_p=g_p/f_p$ and we find

$$\begin{aligned} u_p&=\tfrac{1}{2}[(1-\alpha_p)(1-\beta_p)]^{1/2}\,,\\ v_p&=\tfrac{1}{2}[(1+\alpha_p)(1-\beta_p)]^{1/2}\,,\\ f_p&=\tfrac{1}{2}[(1-\alpha_p)(1+\beta_p)]^{1/2}\,,\\ g_p&=\tfrac{1}{2}[(1+\alpha_p)(1+\beta_p)]^{1/2}\,, \end{aligned} \tag{13}$$

where

$$\alpha_p=(\epsilon_{p-}^2+W^2)^{1/2}/\Omega_p$$

and

$$\beta_p=\epsilon_{p-}/(\epsilon_{p-}^2+W^2)^{1/2}\,. \tag{14}$$

Each operator A_p, B_p, C_p, and D_p destroys one quasiparticle of energy $\Omega_p=(\epsilon_{p-}^2+\Delta_p^2+W^2)^{1/2}$, so the Hamiltonian is diagonal in these quasiparticle coordinates. Note that the energy gap in the excitation spectrum is $(\Delta_p^2+W^2)^{1/2}$ in region 1 and Δ_p in region 2.

We have used the usual BCS mean-field approximation to define Δ_p

$$\Delta_p=\left\langle \sum_{p'n'} V_{pp'nn'}\, c_{-p'n'\downarrow} c_{p'n'\uparrow} \right\rangle\,. \tag{15}$$

The assumed form of $V_{pp'nn'}$ implies that Δ_p is a constant (isotropic) within $\hbar\omega_0$ of the Fermi energy.

IV. THERMODYNAMIC PROPERTIES

Once the transformation to independent quasiparticles is accomplished, it is a simple matter to write down the free energy and calculate the thermodynamic properties. Both the Peierls gap W and the BCS gap Δ are found by minimizing the free energy. Since the model has electron-hole symmetry, the Fermi energy (or chemical potential) is fixed at zero. Then the number of A quasiparticles of momentum p (region 1) is

$$\langle A_p^\dagger A_p\rangle = f(\Omega_p)=1/(e^{\Omega_p/T}+1)\,, \tag{16}$$

with identical results for B, C, and D quasiparticles. We calculate the free energy

$$F(T,\Delta,W)=\langle\mathcal{H}\rangle - TS\,, \tag{17}$$

where for region 1

$$\langle\mathcal{H}\rangle_1=-2\sum_p{}^1 \frac{\epsilon_{p-}^2+W^2}{\Omega_p}\tanh\frac{\Omega_p}{2T} - \frac{V_{\rm BCS}}{N}\left(2\sum_p{}^1\frac{\Delta_p}{2\Omega_p}\tanh\frac{\Omega_p}{2T}\right)^2+\frac{NW^2}{V_p}\,, \tag{18}$$

and the entropy is

$$S_1=-4\sum_p{}^1 f(\Omega_p)\ln[f(\Omega_p)] +[1-f(\Omega_p)]\ln[1-f(\Omega_p)]\,. \tag{19}$$

The momentum sums are taken over momentum space near the Fermi surface of region 1. Equations (18) and (19) reduce to the BCS expressions for $W\to 0$ and in that form would apply to region 2. The present model includes both regions, and so must the sums. The momentum sums are replaced by energy integrals in the usual way:

$$\begin{aligned} \sum_{pn}{}^1 &\to NN_1(0)\int d\epsilon\,,\\ \sum_{pn}{}^2 &\to NN_2(0)\int d\epsilon\,, \end{aligned} \tag{20}$$

where $N_1(0)$ is the electronic density per spin per atom in region 1, and $N_2(0)$ describes region 2 similarly. The total-free-energy per atom is then

$$\begin{aligned} \frac{F}{N}(T,\Delta,W) &= \frac{W^2}{V_p}+\int d\epsilon\sum_l \frac{N_l(0)\Delta^2(\epsilon)}{\Omega_l(\epsilon)}\tanh\frac{\Omega_l(\epsilon)}{2T}\\ &\quad -2T\int d\epsilon\sum_l N_l(0)\ln\left(\frac{\cosh[\Omega_l(\epsilon)/2T]}{\cosh(\epsilon/2T)}\right)\\ &\quad -V_{\rm BCS}\left(\int d\epsilon\sum_l\frac{N_l(0)\Delta(\epsilon)}{2\Omega_l(\epsilon)}\tanh\frac{\Omega_l(\epsilon)}{2T}\right)^2, \end{aligned} \tag{21}$$

where

$$\Delta(\epsilon) = \begin{cases} \Delta, & |\epsilon| < \hbar\omega_0 \\ 0, & |\epsilon| > \hbar\omega_0 \end{cases}$$
$$\Omega_1(\epsilon) = [\epsilon^2 + \Delta^2(\epsilon) + W^2]^{1/2}, \quad (22)$$
$$\Omega_2(\epsilon) = [\epsilon^2 + \Delta^2(\epsilon)]^{1/2},$$

and the ϵ integration is from $-E_B$ to E_B, the electron bandwidth. The constant $F(T, 0, 0)$ has been removed from (21).

The physical values of Δ and W minimize the free energy and are given by the two gap equations

$$W = V_p N_1(0) \int_{-E_B}^{E_B} d\epsilon \, \frac{W}{2\Omega_1(\epsilon)} \tanh \frac{\Omega_1(\epsilon)}{2T}, \quad (23)$$

$$\Delta = V_{\mathrm{BCS}} \sum_I N_I(0) \int_{-\hbar\omega_0}^{\hbar\omega_0} d\epsilon \, \frac{\Delta}{2\Omega_I(\epsilon)} \tanh \frac{\Omega_I(\epsilon)}{2T}. \quad (24)$$

We obtain the following results analytically using the weak-coupling approximation ($kT_m \ll E_B$, $kT_c \ll \hbar\omega_0$). If

$$\frac{1}{N_1(0)V_p} < \frac{1}{N(0)V_{\mathrm{BCS}}} + \ln \frac{E_B}{\hbar\omega_0}, \quad (25)$$

where $N(0) = N_1(0) + N_2(0)$, there are two phase transitions. The martensitic transition occurs first at

$$T_m \simeq 1.134 E_B \exp[-1/N_1(0)V_p], \quad (26)$$

and the superconducting transition follows at a lower temperature given by

$$T_c^{N_2(0)/N(0)} T_m^{N_1(0)/N(0)} \simeq 1.134 \hbar\omega_0 \exp[-1/N(0)V_{\mathrm{BCS}}]. \quad (27)$$

The superconducting transition temperature is reduced by the tetragonality. If, however,

$$\frac{1}{N_1(0)V_p} > \frac{1}{N(0)V_{\mathrm{BCS}}} + \ln \frac{E_B}{\hbar\omega_0}, \quad (28)$$

the superconducting transition occurs at

$$T_c \simeq 1.134 \hbar\omega_0 \exp[-1/N(0)V_{\mathrm{BCS}}], \quad (29)$$

but Eq. (23) cannot be satisfied with $W > 0$, that is, the martensitic transition does not occur.

The following results are obtained by numerically minimizing (21) for parameters appropriate to V_3Si. The susceptibility versus temperature for[11] Nb_3Sn indicates that about 10% of the density of states is removed by the Peierls transition. We assume this number is also appropriate for V_3Si and take $N_1(0)/N(0) = 0.1$. From the observed maximum phonon frequency[12] we take $\hbar\omega_0 = 45$ meV and choose arbitrarily $E_B = 100$ meV. Since we fit actual transition temperatures, our results are quite insensitive to the choice of the cutoff parameters $\hbar\omega_0$ and E_B. The results of this fit (to $T_m = 21$ K and $T_c = 17$ K) are $N_1(0)V_p = 0.242$ and $N(0)V_{\mathrm{BCS}} = 0.283$. We can map out a phase diagram by calculating T_m and T_c vs $N_1(0)V_p$, keeping the other parameters fixed. This is shown in Fig. 1. The phase boundaries are approximately linear in this range of $N_1(0)V_p$ with

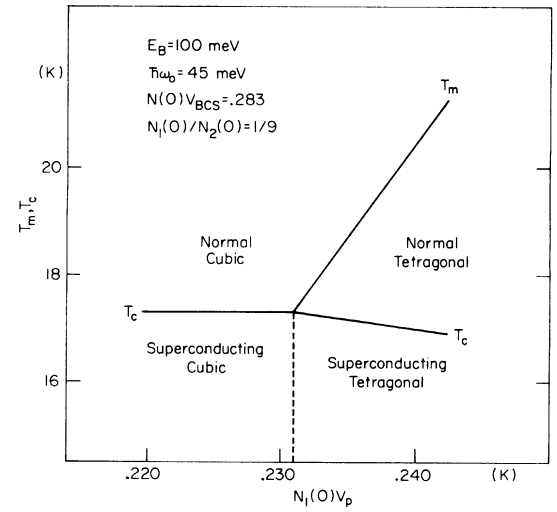

FIG. 1. Theoretical phase diagram obtained by varying the Peierls coupling constant with other parameters fixed as shown.

$$\frac{V_p}{T_m}\frac{dT_m}{dV_p} \simeq \frac{1}{N_1(0)V_p}, \quad (30)$$

and in the tetragonal phase

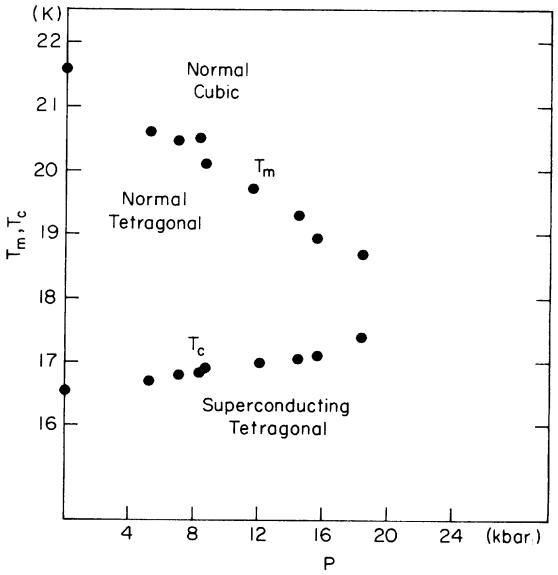

FIG. 2. Experimental phase diagram (Ref. 13) T-P for V_3Si.

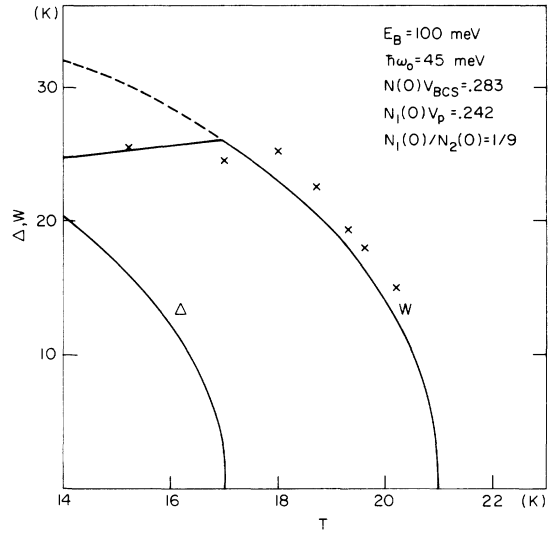

FIG. 3. Calculated plot of $\Delta(T)$, $W(T)$ with parameters appropriate to V_3Si. W continues on dotted path if superconducting transition is prevented. Experimental (Ref. 14) values of $(c/a-1)$, scaled to W below T_c, are given by crosses.

$$\frac{V_p}{T_c}\frac{dT_c}{dV_p} \simeq -\frac{1}{N_2(0)V_p} . \tag{31}$$

These results indicate that $T_c = 17$ K includes a 0.3 K reduction by the open Peierls gap; this effect would be larger if the Peierls gap affected a larger fraction of the Fermi surface. If $N_2(0) = 0$, the slope of T_c is initially infinite but flattens out at higher V_p. It is appropriate to compare this theoretical phase diagram with a plot of transition temperatures versus pressure or versus concentration (in a pseudobinary alloy series). The pressure dependence[13] of T_m and T_c are shown in Fig. 2; it is clearly of great interest to have measurements on V_3Si above 25 kbar. Note that we have held $N(0)V_{BCS}$ constant in the theoretical phase diagram; this means that we have omitted the effects of pressure and of soft phonons on $N(0)V_{BCS}$. The theoretical phase diagram illustrates only the effect of each gap on the other.

The competition of the two gaps on the Fermi surface is clearly shown in the plot of Δ and W versus temperature (Fig. 3), again with parameters appropriate to V_3Si. At the superconducting transition temperature, the growth of the Peierls gap is halted and even slightly reversed as the BCS gap opens up. Since the Peierls gap is directly proportional to the tetragonal distortion, we can compare the measured tetragonality[14] (scaled at low temperature to W) directly with the computed Peierls gap (Fig. 3). Clearly the low-temperature plateau of the distortion is explained by the present model.

V. CONCLUSIONS

We have developed a model for superconductivity and tetragonality in the $A15$ compounds. We have adopted Gorkov's mechanism for the structural or martensitic transition in which a Peierls gap opens up in the one-electron band structure near the X point, and we have included the BCS pair interaction which induces superconductivity. We then solved for the BCS gap and the Peierls gap self-consistently by minimizing the free energy. The interesting new physics that comes out of the model follows from the competition of the two gaps for a common portion of the Fermi surface. The model shows that the Peierls gap (and the tetragonality) is arrested at the superconducting transition (in agreement with experiment) because when both gaps are open Δ and $(\Delta^2+W^2)^{1/2}$ increase essentially identically with falling temperature. The model predicts that the martensitic transition cannot occur at temperatures lower than the superconducting transition temperature. We conclude that where the Peierls gap affects only a small fraction of the Fermi surface, its effect on the superconducting transition is small (0.3 K in V_3Si). Perhaps the most interesting question remaining is the quantitative effect of the soft phonons associated with the martensitic transition on the superconducting transition temperature.

*Research supported in part by NSF contract No. DMR 75-20376.

†Supported by a National Science Foundation fellowship.

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