Strong-Coupling Saturation of Superconductivity in $Nb_3Al_{0.8}Ge_{0.2}$

S. Barišić*

Laboratoire de Physique des Solides,† Université Paris-Sud, Centre d'Orsay, 91405-Orsay, France

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The recent suggestion, based on the tunneling experiments, that the superconducting transition temperature T_c of $Nb_3Al_{0.8}Ge_{0.2}$ is saturated with respect to the variation of the phonon frequencies, is critically examined. This is first done within the McMillan set of ideas concerning the high T_c, which was used in formulating the suggestion. The suggestion is thus shown to be in contradiction with the known pressure dependence of T_c. The observed weak variation of T_c in the martensitic transition is not explained by the saturation effect but rather by the inefficiency of the low-frequency phonons in building up the superconductivity. It is further pointed out that the models of Nb_3X compounds, based on the singularity in the electron density of states, are not expected to yield a saturation of T_c with respect to the phonon frequencies.

In his now classical paper[1] on superconductivity, McMillan has shown that the decrease in the phonon frequencies, characterized, say, by ω_0, leads to an increase in T_c but with an absolute upper limit, which is reached when $\lambda = C/M\omega_0^2$ is roughly $\lambda = 2$. The procedure of maximizing with respect to ω_0 has been carried out while keeping constant the character of the electron system, described by the parameter $C = n_F \langle I^2 \rangle$. Here n_F denotes the bare electron density of states at the Fermi level and $\langle I^2 \rangle$ is an average of the squared electron-phonon matrix element, independent of the phonon spectrum. The maximum of T_c with respect to ω_0 is

$$T_{c\,\max} = (C/2M)^{1/2} e^{-3/2} \tag{1}$$

and depends in particular on C.

McMillan further suggested[1] that such an optimization actually occurs in nature, i.e., that some materials, forming what he calls a "class", have the same C while differing in ω_0. This means, in particular, that within a given class C does not depend on the occupation of the superconducting band. According to Eq. (1), $T_{c\,\max}$ is not constant within a class, due to variation of the average mass M. Apparently, in most cases, McMillan had not considered his estimation to be accurate enough to take care of such a spread in $T_{c\,\max}$. However, he roughly distinguished the mass of the Nb_3Sn-like materials from that of the V_3Si-like materials. In the same way, $T_{c\,\max}$ of $Nb_3Al_{0.8}Ge_{0.2}$ is about 10% larger than that of its heavier classmate Nb_3Sn. Such differences have to be taken into account in estimates claiming an accuracy better than McMillan's.

It was recently suggested,[2] on the basis of the determination of $T_{c\,\max}$ in Nb_3Sn by tunneling experiments, that λ of the high-T_c Nb_3X compounds approach quite closely the $\lambda = 2$ limit; this limit being approached particularly closely for $Nb_3Al_{0.8}Ge_{0.2}$. Clearly, close to this limit T_c will depend only weakly on the variation of ω_0, such as that which may arise from the martensitic transformation. The crystal instability, which may increase ω_0 considerably in the lower-symmetry phase, would therefore not be an important limiting factor in T_c.

This suggestion is analyzed here, first within the McMillan class analysis used in its formulation. By establishing a correspondence between the effects of the critical shear deformation and of the simpler volume compression on T_c, it is shown that the known pressure variation of T_c contradicts the suggestion.

The McMillan equation for T_c establishes a relation between the logarithmic derivatives $\delta_c = \delta \ln T_c$, $\delta_\lambda = -\delta \ln C$, and $\gamma = -\delta \ln \omega_0$, namely[3]

$$(2-\lambda)\gamma = \lambda\delta_c + \delta_\lambda , \tag{2}$$

whatever their origin. In Eq. (2), as well as in Eq. (1), we ignore the Coulomb pseudopotential μ^*, which is generally thought not to be essential for semiqualitative arguments. Equation (2) is valid as long as the relative variations δ, γ are reasonably small. This is the case not only for the usual volume deformations $\delta \ln\Omega$, but also for the critical[4] shear ξ. The possible nonanalytical[5] dependence of δ, γ on ξ does not invalidate Eq. (2), which thus describes the effect on both deformations on T_c.

The conclusion of Ref. 2 amounts to saying that "whatever" γ^ξ is, δ_c^ξ is small, because $|2-\lambda| \ll 1$, while δ_λ^ξ is to be assumed small, in the spirit of the class analysis.

Turning to the noncritical volume deformation, note that here the possibility $|\gamma^\Omega| \gg |\delta_\lambda^\Omega|$ may reasonably be discarded. (This is discussed in more detail below.) With $|2-\lambda| \ll 1$ the term $(2-\lambda)\gamma^\Omega$ can be neglected in Eq. (2), with respect to δ_λ^Ω. δ_c^Ω is then entirely determined by δ_λ^Ω. In other words, the behavior of T_c under pressure should essentially be that of $T_{c\,\max}$, given by Eq. (1).

But while all the McMillan-like interpretations[3,6,8,9] of the pressure data in transition metals and their compounds agree that C, and thus $T_{c\,max}$, increase under pressure p; T_c of $Nb_3Al_{0.8}Ge_{0.2}$ has been shown to decrease.[8] Contrary to what is expected from Eq. (1), δ_c^Ω and δ_λ^Ω are of the same sign; thus $|2-\lambda|$ in the positive $(2-\lambda)\gamma^\Omega$ term must be large enough to satisfy the equality (2). (Two somewhat different roles may be ascribed to the γ term: It can cancel out the leading $\delta \ln\Omega$ term in δ_λ^Ω and yield the recently proposed[7] quadratic dependence of $\delta \ln T_c$ on $\delta \ln\Omega$. It can also overwhelm this term and give the correct sign to the linear dependence of $\delta \ln T_c$ on $\delta \ln\Omega$, claimed by other authors.[8])

Equation (2) may serve for a determination of λ, if instead of the poorly known δ^ℓ, γ^ℓ, the better known $\delta^\Omega, \gamma^\Omega$ are used. In what follows, we adopt the usual[3] definition of $\delta^\Omega, \gamma^\Omega$, i.e., we divide the quantities δ, γ by the volume deformation $\delta \ln\Omega$. Note that $\delta \ln\Omega = -p/K$, K being the bulk modulus, is negative. First, let us briefly discuss the quantities involved in Eq. (2), starting with δ_c^Ω.

Concerning δ_c^Ω, the value of $\delta T_c/\delta p \times 10^{-5}$ °K bar^{-1}, measured[8] up to 25 kbar, is -1.4 and -0.2 for Nb_3Sn and $Nb_3Al_{0.8}Ge_{0.2}$, respectively. For Nb_3Sn this corresponds to $\delta_c^\Omega \approx 1.25$, while the value quoted for $Nb_3Al_{0.8}Ge_{0.2}$ should rather be considered[8] as corresponding to an upper limit of δ_c^Ω.

The typical values of δ_λ^Ω are close to 2.[3] This can be understood on applying the tight-binding argument[6,9] to the McMillan C, which is to be considered independent of the band filing. Excluding, therefore, the possibility of the contribution to δ_λ^Ω from the charge transfer among the s-p and d bands we have, from Refs. 6 or 9, $\delta_\lambda^\Omega \approx 1$. This is of the correct order of magnitude. [The terms of the higher order in $\delta \ln\Omega$ are generally negligible with respect to the leading one retained here. Thus, the appearance of the higher-order terms in δ_c^Ω in this model only results from such terms in γ^Ω, after a (partial) compensation[7] of the leading terms among δ_λ^Ω and γ^Ω].

The coefficient γ^Ω is similar in nature to the Grüneisen constant $\gamma_G(k,\nu)$. For long-wavelength (l-w) shear phonons, γ_G(l-w, ξ) was found[10] to be negative. This can be understood from the pressure enhancement of the phonon frequency renormalization, arising from the same effect[6] as the positive δ_λ^Ω. In the case of V_3Si, this led to the proposals that $\gamma^\Omega \approx 0$[9] or $\gamma^\Omega < 0$.[7] However, the tunneling data do not attribute any special role to the extremely long-wavelength shear phonons, in agreement with the previous theoretical suggestion[6,11] that due to the retardation effects the (critical) phonons with $\omega \lesssim 2k_BT_c$ (≈ 3 meV in Nb_3Sn) do not contribute significantly to λ, whatever it is. The electron renormalization of the lowest important phonons, at 9 meV in Nb_3Sn, is considerably weaker[12,13] than in the extreme l-w limit, even for the shear branch. It thus seems reasonable to think that $0 < \gamma^\Omega < \gamma_{G\,th}^\Omega$, where $\gamma_{G\,th}^\Omega$ is the high-temperature thermal Grüneisen constant, typically[10] equal to 1.5 for the compounds under consideration. However, only the upper limit on γ^Ω is important for the conclusion which follows; the weaker assumption $|\gamma^\Omega| < \gamma_{G\,th}^\Omega$ works as well.

The conclusion is that both Nb_3Sn and $Nb_3Al_{0.8}Ge_{0.2}$ fall rather far from the saturation limit. Equation (2), with $\delta_\lambda^\Omega \approx 2$ and $|\gamma^\Omega| \lesssim 2$, yields for Nb_3Sn $2-\lambda \gtrsim 1.5$ if $\gamma^\Omega > 0$ and $\lambda - 2 \gtrsim 6$ if $\gamma^\Omega < 0$. For $Nb_3Al_{0.8}Ge_{0.2}$ it gives roughly $|2-\lambda| \gtrsim 1$. The unphysically large values of λ, associated with $\lambda^\Omega < 0$, together with the above-mentioned argument against $\gamma^\Omega < 0$ itself, suggest that this possibility does not occur in the present case. These conclusions remain essentially unchanged when the Coulomb pseudopotential μ^* is taken into account, especially as far as the estimate of the distance from the saturation value of λ is concerned.

It should be noted that the above type of reasoning cannot be extended to the materials with $\delta_\lambda^\Omega \ll -\lambda\delta_c^\Omega$ and thus to V_3Si and V_3Ge, which have a large and negative δ_c^Ω. Unlike the favorable situation discussed above, Eq. (2) is then of no help in putting the limits on $|2-\lambda|$ or in choosing the sign of γ^Ω.

However, it seems well established now[14,15] that T_c is roughly the same in the cubic (nonsheared) and tetragonal (sheared) phase of Nb_3Sn, although the Grüneisen constant with respect to the shear ζ, $\gamma_G^\zeta(k,\xi)$ is large for the l-w shear phonons, both experimentally[17] and theoretically.[6,10,16] Such behavior of T_c is not explained here by $\lambda \approx 2$ arguments but, rather, consistently with the unimportance of the large negative γ_G^Ω(l-w, ξ), by the exclusion[6,12] of the l-w shear phonons with $\omega \gtrsim 2k_BT_c$ from λ, and thus from γ^ℓ. Still, T_c may be further increased by decreasing ω_0 through other phonons.

It is finally argued that these conclusions remain essentially correct for the models based on the sharp peak in the density of electron states, and currently used for explaining the crystal instability. With the Fermi level close to sufficiently isolated Van Hove points[6,11] on the energy surface, λ equals $n_F V^{sp}$ with V^{sp} ($\sim \omega_0^{-2}$) possibly dependent on the band filling, but only through the dependence of ω_0, which is already described by γ. However, because of the factor n_F, such a λ is strongly dependent on the band filling, i.e., on the charge transfer s-$p \rightarrow d$. The McMillan class analysis has to be abandoned. But a new possibility appears — that of saturation with respect to the band occupation at "constant" V^{sp} — and does occur[17,18] in the Labbé-Friedel model,[19] while the saturation with respect to ω_0 is pushed to the

large, generally unphysical values of λ. In an alternate model,[20] which is much less singular than the Labbé-Friedel one, this latter saturation occurs for $\lambda \approx 4$ provided that the corresponding ω_0 is still larger than E_F counted with respect to the singularity. The evidence[21] for the pronounced peaks of T_c in the ternary $A_3B_xB_{1-x}$ compounds, such as the peak[21] at $Nb_3Al_{0.8}Ge_{0.2}$, agrees with the idea of a maximum of such a nature, since the variation of x leads, in the first place, to the variation of the band occupation. The pressure dependence of the $T_{c\,max}$ so defined was previously examined[8,9] and led to an equation similar to Eq. (2) taken far below the $\lambda = 2$ limit. Everything that was said above about such an equation for the McMillan analysis thus remains qualitatively unaltered.

Obviously, further careful measurements and analyses of both the pressure dependence of T_c and the tunneling characteristics, are necessary to resolve the discrepancy between the values of λ deduced from the two types of data. These analysis should be combined with the neutron determination of the phonon density of states $F(\omega)$, in order to distinguish between the electron and the phonon features of the tunneling function[1,2] $\alpha^2(\omega)F(\omega)$, since certain classes of phonon may be favored[6,11] by the anisotropy of the electron spectrum. The possibility that the 9-meV anomaly observed[2] in the function α^2F of Nb_3Sn is of such a nature and is currently under theoretical investigation.

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*Present address: Institute of Physics, University of Zagreb, Croatia, Yugoslavia.

†Laboratoire associé au Centre Naciónal de la Recherche Scientifique.

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