# Electron tunneling into $Nb_3Sn$, $Nb_3Ge$, and $Nb_3Al$

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We have prepared high-quality tunnel junctions on thin films of $Nb_3Sn$, $Nb_3Ge$, and $Nb_3Al$ with artificial tunnel barriers of Al oxide and AlZr oxide. The $Nb_3Sn$/AlZr-oxide/Pb tunnel junctions prepared on Sn-deficient material showed no measurable proximity effect. Whereas the tunnel spectra of Sn-deficient $A15$ NbSn could be satisfactorily analyzed by the standard Rowell-McMillan inversion procedure, the reduced density of states of the stoichiometric samples exhibited characteristic deformations in the form of an overswing at energies far above the highest phonon energy. These deformations can be fitted by proximity analysis, but it is shown that they can also be referred to the influence of an electronic density of states which is strongly structured in the range of phonon energies near the Fermi level. A very similar behavior is found for $Nb_3Ge$ and to a smaller extent for $Nb_3Al$.

## I. INTRODUCTION

The highest superconducting transition temperatures $T_c$ have been found so far in materials of $A15$ crystalline structure. Therefore, these materials have been extensively investigated both experimentally and theoretically. In the theory of superconductivity, the quantity of central interest is the Eliashberg function $\alpha^2(\omega)F(\omega)$ which describes Fermi-surface scattering of quasiparticles via the emission and absorption of phonons and is defined as the product of the phonon density of states $F(\omega)$ and an energy-dependent electron-phonon coupling function $\alpha^2(\omega)$. For some $A15$ and $B1$ superconductors, $\alpha^2F$ as well as the phonon density of states have been calculated by Weber[1] using the non-orthogonal tight-binding method based on realistic energy-band calculations. Weber[1] finds that in the high-$T_c$ superconductors of both material groups, there are phonons which are strongly softened by an enhanced electron-phonon coupling strength giving rise to an increased $\alpha_2(\omega)$ in the corresponding energy region. For $Nb_3Sn$, an $A15$ superconductor which has been studied most extensively in our work, these soft modes are located in a low-energy peak in $\alpha^2F$ near 8 meV. Another interesting feature typical for the $A15$ materials is that following the energy-band results,[2] these materials have an electronic density of states (EDOS) which is strongly structured on the scale of typical phonon energies in the vicinity of the Fermi edge. In this case the Eliashberg equations as used so far in tunneling analysis are not valid. Kieselmann and Rietschel[3] and independently Mitrovic and Carbotte[4] have solved the Eliashberg equations explicitly with the inclusion of a rapidly varying EDOS and found characteristic deformations in the reduced tunneling density of states (RDOS). Most evident is an enhanced overswing of the RDOS data far above the highest phonon energy. An attempt to detect these deformations in a tunneling experiment can only be successful if the junctions are free from other deformations such as those due to proximity effects. So far all tunneling results published for high-$T_c$ superconductors suffered from proximity effects.[5–7] In these cases the tunneling barriers used were natural oxides or oxides of thin Si overlayers deposited *in situ* on a freshly prepared thin film of a superconductor. In this paper, we present an advanced preparation technique based on overlayers of Al and AlZr which allow for proximity free junctions on some $A15$ superconductors.

## II. EXPERIMENTAL

The $Nb_3Sn$ and $Nb_3Al$ tunneling samples were prepared by magnetron sputtering. The sputtering targets of the superconductor and of the overlayer (Al or AlZr) were mounted on a big cathode plate of a 15-cm diameter. After deposition of the superconductor, an external ring magnet was shifted to the overlayer target. During deposition of the $A15$ thin films, substrate temperatures near 1000 °C were used. The $Nb_3Ge$ tunneling samples were prepared in a multiple electron-gun system as described in Ref. 8. All of the junctions prepared with overlayer technique were oxidized not longer than 10 min in laboratory air for the formation of the tunnel barrier oxide. A detailed description of the preparation technique of our tunneling samples will be published elsewhere. All the tunneling samples of this study were characterized by x-ray and Rutherford backscattering measurements. The derivative measurements were taken by standard first and higher harmonic detection and on-line computer data collection.

## III. RESULTS AND DISCUSSION

In Fig. 1 we show a $dI/dV$ versus $V$ trace of a $Nb_3Sn$/AlZr-oxide/Pb junction with the Pb counterelectrode in the superconducting state. We observe that for voltages below the lead gap (1.37 meV), the junction isolates to better than 1 part in $10^3$. Above the lead gap till about 4 meV, the junction exhibits a trace similar to a

 

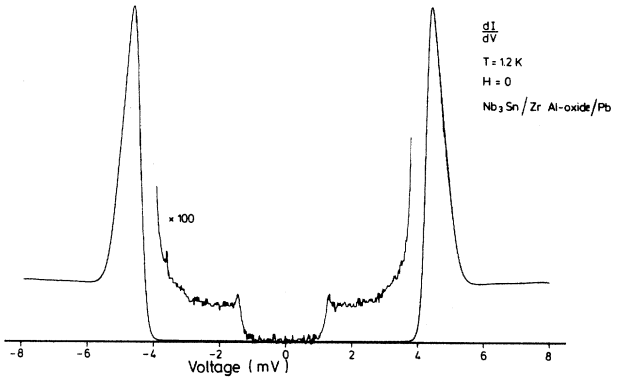


FIG. 1. Direct reproduction of the $dI/dV$ recorder trace of a $Nb_3Sn$/AlZr-oxide/Pb junction taken at 1.2 K with both electrodes in the superconducting state.

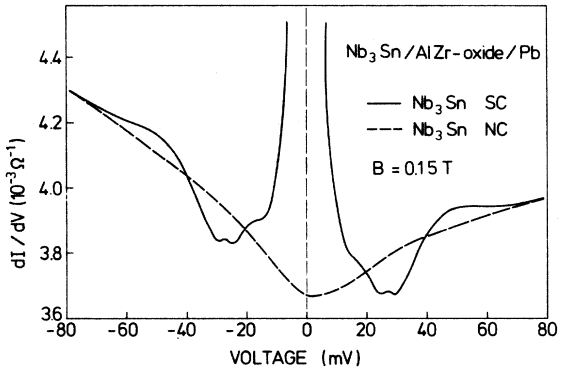


FIG. 2. Derivative measurements ($dI/dV$ vs $V$) for an optimally prepared $Nb_3Sn$/AlZr-oxide/Pb tunnel junction with the $Nb_3Sn$ in both the superconducting and normal states. The superconductivity of the Pb counterelectrode is suppressed by a magnetic field.

Pb/isolator/normal-conductor junction. This tunnel current is therefore interpreted as being due to tunneling into a small amount (about 0.7% in this case) of normal grains present at the film surface. The peaks at the sum of the gaps near $\pm 4.5$ meV show a finite width of about 0.7 meV. The most simple explanation is a corresponding spread of energy gaps in the $Nb_3Sn$ film. By carefully analyzing the opening of the energy gap near $T_c$, we found that there is also a corresponding $T_c$ spread in the samples. Thus, we exclude gap anisotropy as the main cause for the finite width of the peaks at the sum of the gaps. For the calculation of the RDOS which represents the input data of the inversion program of McMillan and Rowell (MMR), the tunnel data were corrected for the excess currents below the sum of the gaps as described in Ref. 8.

Junctions prepared with different types of tunnel barriers (natural oxides of the $A15$ film, Al oxide, AlZr oxide) showed the same characteristic improvement which we have reported for artificial Al-oxide barriers on Nb.[9] For $Nb_3Sn$ we obtained optimum results with AlZr-oxide barriers. In this case similar to the data reported in Ref. 9 the rapid increase of the normal-state $dI/dV$ data appears at $Nb_3Sn$ positive bias. For nonoptimum junctions, where Nb oxides contribute to the tunnel barrier, the rapid increase is at negative bias. In Fig. 2 we show $dI/dV$ data (the "raw data" of the tunneling experiment) for an optimally prepared $Nb_3Sn$ tunnel junction. We see that the data with $Nb_3Sn$ positive bias show a very flat behavior and are best suited for tunneling spectroscopy.

In Fig. 3(b) we show RDOS results for $Nb_3Sn$/AlZr-oxide/Pb junctions with an Sn content of 19 at.% ($T_c = 7$ K) and in Fig. 3(a) for a stoichiometric high-$T_c$ sample with $T_c = 18$ K and the fits obtained by McMillan and Rowell and the proximity McMillan-Rowell (PMMR) analysis. We see that the RDOS data of the Sn deficient sample could be inverted very well using the standard MMR analysis except for a slight deviation between 40 and 60 meV. The resulting $\alpha^2F$ is shown in Fig. 3(c). For the high-$T_c$ RDOS data, no acceptable fit was obtained by MMR analysis due to the conspicuous overswing of the RDOS data between 40 and 60 meV. Therefore, we tried the proximity-inversion analysis as introduced by Arnold.[10] In this analysis the experimenter tries to get a fit to the measured RDOS data by the appropriate adjustment of two additional parameters, $d/l$ and $R = 2d/(hv_F)$ where $d$ denotes the thickness of a proximity layer between the superconductor and the tunnel barrier, $l$ is an effective mean free path in this layer and $v_F$ is its Fermi velocity. In Fig. 3(a) we show the best fit we obtain to our high-$T_c$ RDOS data by PMMR and in Fig. 3(c) we show the resulting $\alpha^2F$ function. For the proximity parameters we get $d/l = 0$ and $R = 0.013$ (meV)$^{-1}$. The $R$ parameter describes Andreev oscillations in the proximity layer and its comparably high value takes care of the enhanced overswing between 40 and 60 meV in our high-$T_c$ data. Tunnel measurements on samples with Sn contents between those of Fig. 3 show that the enhanced overswing feature is continuously increasing from low- to high-$T_c$ material.

There are several arguments which made us doubt that a proximity effect is the cause of the enhanced overswing in our high-$T_c$ RDOS data of $Nb_3Sn$: (i) The result that $d/l = 0$ and $R$ takes a comparably high value. To our knowledge such a set of proximity parameters has not been found so far on proximity junctions. For example, for Nb junctions, one finds a decrease of both parameters with increasing junction quality.[9] (ii) We found the enhanced overswing always occurring with the same shape and in the same energy region independently of the special tunnel barrier used (natural oxides, very thin and thick Al overlayers, AlZr overlayers) and independently of the strength of the proximity effect. It seems very unlikely that different types of tunnel barriers with a different structure of the barrier-superconductor interface should result in the same value of $d/v_F$ of that interface. (iii) On high-$T_c$ junctions exhibiting proximity effects ($d/l > 0$ from PMMR analysis) we always found dips above the sum of the gaps in the $dI/dV$ curves. In the $I$-$V$ curves of superconductor-insulator-superconductor (SIS) proximity junctions these dips are reflected as the typical "knee" structures. On our optimally prepared junctions, these dips are not present (see Fig. 1). We remind the reader than on a SIS junction with a Pb counterelectrode ($\Delta_0 = 1.37$ meV), much smaller dip structures are visible

than with, for example, In counterelectrodes ($\Delta_0=0.53$ meV) as used by Wolf *et al.*[11] (iv) We do not see why a proximity layer should only be present in junctions on high-$T_c$ and not on low-$T_c$ material. In fact, we found that on preparation runs which were not carried out under optimum conditions, the junctions showed the undesirable proximity properties ($d/l>0$, "knee" structures) independently of stoichiometry and $T_c$ of the samples.

In the following, we present an alternative way to explain the enhanced overswing feature of our high-$T_c$ junctions. It is given by the work of Kieselmann and Rietschel (KR) (Ref. 3) who included a strongly structured EDOS into the Eliashberg equations and found that as a consequence the RDOS is deformed mainly in two ways: (i) An overall increase of the RDOS in magnitude and (ii) an enhanced overswing. In nonperfect crystals, for example, in materials deviating from stoichiometry, a smearing and decrease of the EDOS structure is theoretically expected[12] and has been found experimentally by electron energy-loss spectroscopy.[13] In terms of the KR theory this would explain the disappearance of the enhanced overswing on our Sn deficient samples. The specific residual resistivity of our high-$T_c$ samples was near 30 $\mu\Omega$ cm and increased to about 130 $\mu\Omega$ cm for the Sn deficient sample with a $T_c$ of 7 K. Following the results of Pickett,[12] even with 30 $\mu\Omega$ cm a smearing of the EDOS structure can be expected. But we remind the reader that tunneling takes place into the single crystalline grains of the film which for stoichiometric material may have a much smaller resistivity compared to the overall value of the thin film which includes scattering at the grain boundaries and in the first few hundred angstroms where the deposition has started and which are usually more contaminated.

In a first attempt to analyze our data on the basis of the KR work we used the KR program (the program calculates the RDOS from a given $\alpha^2F$ and EDOS) with structured EDOS curves and an $\alpha^2F$ composed of three peaks of Lorentzian shape which were adjusted such that the re-

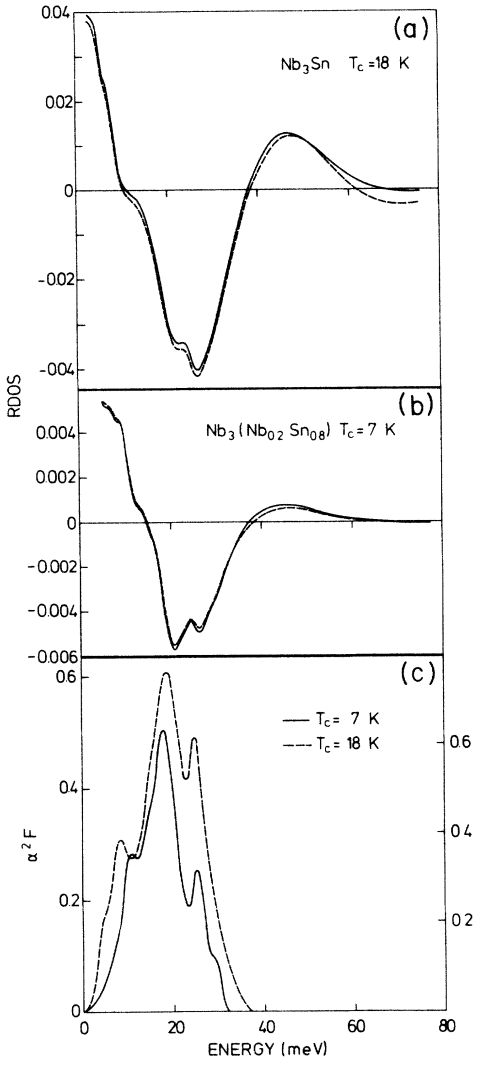


FIG. 3. (a) PMMR analysis of high $T_c$ $Nb_3Sn$. The experimental RDOS data (solid line) were fitted by PMMR (dashed line) with the proximity parameters $d/l=0$, $R=0.013$. (b) MMR analysis of low $T_c$ $A15$ NbSn. Experimental data, solid line; calculated fit, dashed line. (c) Eliashberg function resulting from the analysis. Low $T_c$ $A15$ NbSn: solid line, left scale. High $T_c$ $A15$ $Nb_3Sn$: dashed line, right scale. The parameters are for low-$T_c$ $A15$ NbSn: $d/l=0$, $R=0$ (MMR), $\lambda=0.9$, $\mu^*=0.26$, $\Delta_0=0.93$, $\langle\omega\rangle=14.6$ meV, $\langle\omega^2\rangle=255$ (meV)$^2$. For high $T_c$ $Nb_3Sn$: $d/l=0$, $R=0.013$ (PMMR), $\lambda=1.84$, $\mu^*=0.33$, $\Delta_0=3.15$, $\langle\omega\rangle=14.0$ meV, $\langle\omega^2\rangle=255$ (meV)$^2$.

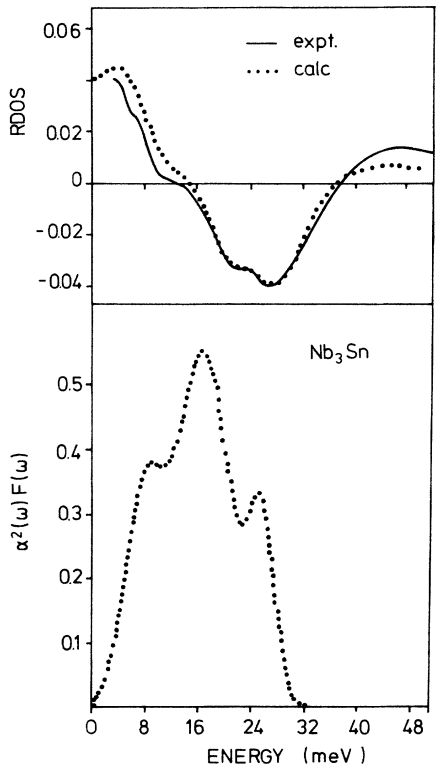


FIG. 4. Upper graph: The solid line shows the measured RDOS of a high $T_c$ $Nb_3Sn$/AlZr-oxide/Pb junction ($T_c=18$ K) and a fit calculated by the KR program using as input a parametrized $\alpha^2F$ (lower graph) and the EDOS of Ref. 2. Lower graph: The $\alpha^2F$ function for the RDOS fit. $\langle\omega\rangle=12.4$ meV.

sulting RDOS approximately fits our experimental data. The upper part of Fig. 4 shows the experimental RDOS curve along with a fitted curve which was obtained by the $\alpha^2F$ shown in the lower part of Fig. 4. In this calculation we used the EDOS curve calculated for $Nb_3Sn$ by Mattheiss and Weber[2] which shows one sharp peak above the Fermi edge near 20 meV. We see in Fig. 4 that the steep rise of the experimental data between 27 and 35 meV is reproduced well but the calculated curve shows a smaller overswing than the measured one. In Fig. 5 we show a fit calculated with an $\alpha^2F$ of a shape very similar to that of Fig. 4 but using an EDOS curve with two peaks located above and below the Fermi edge. We see that the resulting RDOS follows the overswing feature of our measured data very closely. In the course of the KR analysis we found that the shape of the resulting $\alpha^2F$ did not change much on using different EDOS curves. But the $\alpha^2F$ varied strongly in scale and so did the resulting superconducting parameters, too. As we still do not know if there exists an unequivocal solution of our measured data with respect to the EDOS we do not present here the superconducting parameters $\mu^*$ and $\lambda$ and EDOS curves resulting from our preliminary KR analysis. We only point out that it is possible to describe our high-$T_c$ $Nb_3Sn$ data quantitatively using an $\alpha^2F$ of reasonable shape in connection with a structured EDOS without need for any further corrections (e.g., with respect to proximity effects). We mention that a KR analysis is only possible on tunnel data which exhibit $d/l=0$ on proximity analysis. This is due to the depression of data with $d/l>0$ into negative direction resulting in RDOS data which deviate from particle number conservation.[14] But MMR as well as KR analysis require RDOS data with particle number conservation.

It is of interest if the enhanced overswing feature can be found in other $A15$ superconductors, too. In Fig. 6 we compare RDOS data for high- and low-$T_c$ material of $Nb_3Al$ and $Nb_3Ge$. In order to facilitate the comparison, the RDOS of the Al and Ge deficient samples have been rescaled to match the strength of the high-$T_c$ data in the energy region of the phonons. For $Nb_3Ge$ we see a behavior strongly resembling that of $Nb_3Sn$, i.e., a clearly enhanced overswing for the high-$T_c$ sample. In these studies we obtained only $Nb_3Ge$ samples with a $T_c$ up to 17.5 K. Kihlstrom *et al.*[15] demonstrated that it is possible to prepare $Nb_3Ge$ junctions with a $T_c$ up to 21 K and it would be interesting to pursue the further increase of the overswing feature. A preliminary proximity analysis of our $Nb_3Ge$ samples which was carried out only till 40 meV showed an acceptable quality of the samples with $d/l$ values near 0.02. In Fig. 7 we compare our high-$T_c$ $Nb_3Ge$ data taken on a $Nb_3Ge$/Al-oxide/In junction to those by Schmidt *et al.*[16] for a $Nb_3Ge$/$Nb_3Ge$-oxide/Pb junction (the first RDOS data published for $Nb_3Ge$). In the low-energy region, this junction shows the depression of the RDOS data into negative direction typical for proximity effects, which so far was always observed for junctions with tunnel barriers composed of Nb oxide. We see in Fig. 7 that both junctions show the same enhanced

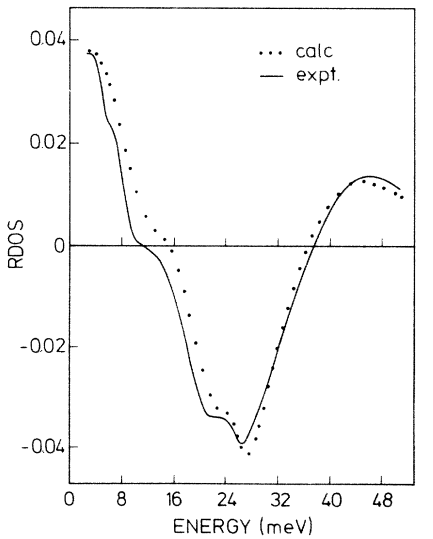


FIG. 5. Experimental RDOS of Fig. 4 (solid line) and a fit calculated by the KR program using as input an $\alpha^2F$ similar to Fig. 3(a) and an EDOS containing a peak above and a peak below the Fermi edge. The purpose of the figure is to demonstrate that there exists at least one set of $\alpha^2F$ and EDOS functions to explain the overswing of the experimental data above 40 meV within the KR theory.

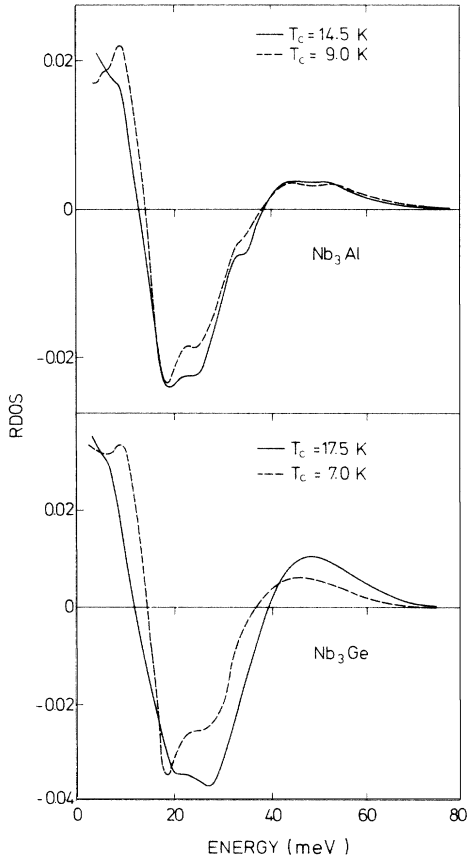


FIG. 6. Comparison of the RDOS data of high and low $T_c$ $A15$ materials ($Nb_3Al$, upper graph; $Nb_3Ge$, lower graph). The scaling is valid for the high-$T_c$ traces. The traces of the low-$T_c$ samples have been rescaled to match the strength of the high-$T_c$ RDOS in the energy region of the phonons in order to facilitate the comparison.

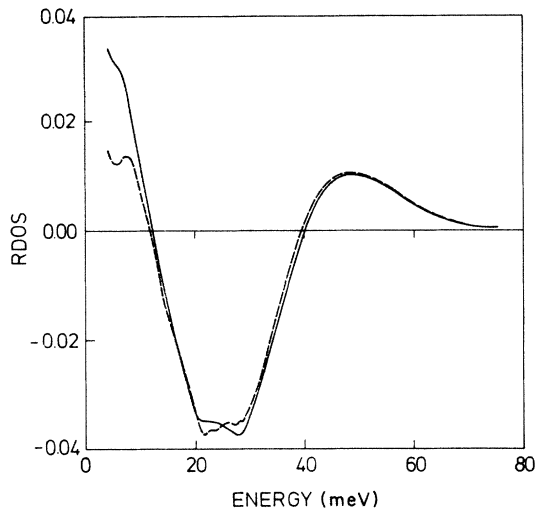


FIG. 7. Comparison of RDOS data from a $Nb_3Ge$/Al-oxide/In junction ($\Delta_0=3.25$ meV, solid line) to data from a $Nb_3Ge$/$Nb_3Ge$-oxide/Pb-junction ($\Delta_0=3.4$ meV, dashed line) (see Ref. 16).

overswing despite the obviously different structure of their barrier-superconductor interfaces. The same findings were mentioned above for $Nb_3Sn$ as an argument ruling out proximity effects as a cause for the enhanced overswing.

For $Nb_3Al$ (Fig. 6) we obtained junctions with a maximum $T_c$ of only 14 K. This may be the reason that the overswing increase shows up only very weakly for these junctions. A proximity analysis of these data yields $d/l=0$, $R=0.001$ for the low-$T_c$ and $d/l=0$ and $R=0.010$ for the high-$T_c$ $Nb_3Al$ sample. Taking the $R$ parameter as an indicator, we believe that also for high-$T_c$ $Nb_3Al$ there is a slightly enhanced overswing hidden in the RDOS. A structured EDOS for $Nb_3Al$ has been proposed by Junod *et al.*[17] to explain susceptibility measurements. Kwo and Geballe[7] prepared tunnel diodes on high-$T_c$ (14.0—16.4 K) $Nb_3Al$ using tunnel barriers of Si oxide. The diodes were proximity affected showing $d/l=0.06$ on PMMR analysis. Except for a depression into negative direction typical for proximity effects, their high-$T_c$ data are similar to ours.

The proximity parameters resulting from PMMR analysis of our low-$T_c$ $A15$ NbAl diodes show that the data are very close to the ideal case of MMR behavior as we achieved for low-$T_c$ $A15$ NbSn. The resulting $\alpha^2F$ data show near 35 meV a peak which is about a factor of 12 smaller than expected from the phonon density of states. This peak reflects the almost localized vibrations of the Al atoms and our results suggest a very small coupling of the electrons to these phonons. More details will be given in a forthcoming paper.

## IV. SUMMARY AND CONCLUDING REMARKS

Using magnetron sputtering we have developed an overlayer technique for the preparation of tunnel junctions with artificial tunnel barriers of Al and AlZr oxide. For $Nb_3Sn$ we have shown that in this way proximity-free tunnel junctions can be fabricated on Sn-deficient material. Junctions prepared on high-$T_c$ $Nb_3Sn$ showed RDOS data which could not be inverted by the usual McMillan-Rowell program due to an enhanced overswing at energies above the highest phonon energy. We have listed several arguments against proximity effects to generate this feature. We presented an analysis (KR) based on Eliashberg equations containing a strongly structured EDOS and explained our tunnel data without the need of further corrections for proximity effects. As the EDOS structures are expected to smear out with increasing disorder, the disappearance of the enhanced overswing feature on the Sn-deficient samples can also be explained in terms of this analysis. Of course, there are other possibilities which could generate the observed anomaly in the RDOS. One is anisotropy effects. We studied our samples by scanning-electron microscopy and found that the surface of the individual grains is rounded off, so that there are many different crystalline planes covered by the tunnel barrier and the counter electrode. We do not expect anisotropy effects to arise solely from the surface structure of our films. But it is conceivable that the overlayers as well as the oxides grow in a slightly different way on the different planes. As only a one-Angstrom change in the thickness of the tunnel barrier causes about an order-of-magnitude change in the tunneling resistance, this could provide a strong selectivity of the tunneling experiment to those crystalline directions where the tunnel barrier is the thinnest. Such a "masking" effect of the tunnel barrier could be revealed simply by preparing the junctions in such a way that the superconductor to be investigated forms the counterelectrode, as has been done for Pb and other simple metals. For $A15$ superconductors this is technically impossible.

Of course, additional information on the high-$T_c$ $A15$ materials is needed, for example, EDOS measurements with very high resolution, to definitively check the two proposed models (PMMR and KR). At present we can only give scenarios following from these models. (i) If the enhanced overswing feature is really caused by proximity effects, then the $\alpha^2F$ and the parameters given in Fig. 2 describe high-$T_c$ $Nb_3Sn$. From the $\langle\omega\rangle$ values we conclude that no mode softening is responsible for the high $T_c$ in $Nb_3Sn$. This is due to the high-$R$ value needed to closely describe the overswing and which blows up the high-energy part of the $\alpha^2F$, but is in contradiction to published PMMR results of $Nb_3Sn$.[5] We attribute this disagreement to two facts. First, due to the Al-oxide barriers, our junctions are of higher quality. Second, we have measured and analyzed our tunneling data on a wider range of energies, down to lower energies and more important by up to much higher energies, completely covering the overswing. The data published so far for $Nb_3Sn$ (Ref. 5) and $Nb_3Ge$ (Ref. 6) were measured and analyzed only up to 40 meV, i.e., where the overswing just starts. (ii) If the KR analysis turns out to be the correct description (in the sense that the enhanced overswing feature is due to a structured EDOS), then high-$T_c$ $Nb_3Sn$, $Nb_3Ge$, and $Nb_3Al$ have to be described by modified Eliashberg equations (KR), perhaps also the applicability of Migdal's theorem in this case has to be checked over again. Our

$\alpha^2F$ result for $Nb_3Sn$ from the KR analysis is somewhat similar to the published PMMR results[5] and shows also some mode softening with respect to our low-$T_c$ results. The extent to which this mode softening causes the high $T_c$ can only be checked by Eliashberg equations containing the correct EDOS function.

In conclusion we state that, at present, reliable $\alpha^2F$ data of the $A15$ superconductors $Nb_3Sn$, $Nb_3Ge$, and $Nb_3Al$ can only be determined by MMR analysis of high-quality diodes prepared on low-$T_c$ material, for example, 7 K for $Nb_3Sn$. Unfortunately, most of the existing phonon density-of-states data to which the $\alpha^2F$ data should be compared were taken on the more attractive high-$T_c$ stoichiometric samples. Due to difficulties in the calculation of the band structure, theoretical data on electron-phonon coupling in off-stoichiometric $A15$ materials are very rare. Therefore it is important to clarify completely the physical nature of the overswing behavior of the high-$T_c$ RDOS data and to develop an adequate inversion procedure for the determination of the Eliashberg function.

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