PHYSICAL REVIEW B VOLUME 32, NUMBER 5 1 SEPTEMBER 1985

Properties of superconducting vanadium nitride sputtered films

John Zasadzinski
Illinois Institute of Technology, Chicago, Illinois 60616

R. Vaglio and G. Rubino
Dipartimento di Fisica, Università degli Studi di Salerno, I-84100 Salerno, Italy

K. E. Gray
Materials Science and Technology Division, Argonne National Laboratory, Argonne, Illinois 60439

M. Russo
Istituto di Cibernetica del Consiglio Nazionale delle Ricerche, I-80072 Arco Felice, Italy

(Received 15 April 1985)

High-quality films of VN were prepared by reactive dc sputtering with the use of a triode source. The films were characterized by x-ray diffraction, temperature-dependent electrical resistivity, and superconducting T_c. Deposition onto single-crystal sapphire substrates heated to 600 °C resulted in uniaxial growth of grains along the [111] direction. Residual resistance ratios as high as 5 were obtained and T_c up to 8.9 K. The dependence of T_c on a lattice parameter differs considerably from previous reports in the literature. The temperature-dependent resistivity was found to be consistent with the parallel-resistor model. Tunnel junctions were fabricated by direct plasma oxidation of the film surface and preliminary quasiparticle results confirm that VN exhibits strong electron-phonon coupling. Josephson junctions utilizing an artificial CdS barrier display diffraction characteristics leading to an upper-bound estimate of the penetration depth of $\lambda \sim 1300$ Å.

I. INTRODUCTION

The $3d$ transition-metal nitrides have long been of interest due to their unusual physical properties, which are of technological as well as fundamental importance.[1] Strong interest in VN has been stimulated recently by Rietschel *et al.*,[2] who argue that spin fluctuations (i.e., paramagnons) reduce the T_c of VN from ≈ 30 K to the observed bulk value of about 9 K. Earlier predictions[3] indicated a smaller suppression of T_c in Nb and V, and recently,[4] tunneling spectroscopy data on Nb, V, and Ta have been reanalyzed using a rescaled form of the Eliashberg equations which was demonstrated by Daams *et al.*,[5] to account for paramagnon effects. It was concluded[4] that T_c in V is indeed suppressed by up to 45%. However, the larger magnitude estimated[2] for VN makes it a natural choice to study paramagnon effects in detail. Spin fluctuations (paramagnons) are the precursor of band ferromagnetism, and as such they suppress the antiparallel alignment of electron spins in a Cooper pair. Although the paramagnon spectral density has not been observed directly, a measure of the potential influence of spin fluctuations is the exchange enhancement of the spin susceptibility, given by the Stoner factor. Another indication of paramagnon effects is the contribution to the electron mass enhancement in heat capacity or de Haas—van Alphen. In such measurements the mass enhancement is $m^*/m = 1+\lambda_p+\lambda_s$, where λ_p and λ_s are, respectively, the electron-phonon and electron-spin fluctuation coupling strengths. Note that while the coupling strengths add in m^*, they have opposite effects for superconductivity since λ_s is a repulsive electron-electron interaction like the Coulomb interaction (μ^*). Roughly speaking, the net attractive interaction is $\lambda_p - \lambda_s - \mu^*$, so that if $\lambda_s > \lambda_p - \mu^*$, there can be no superconductivity. Thus, the large values of S and λ_s in Pd and Sc are consistent with the absence of superconductivity in these elements.[3,6] It has also been suggested[7] that the dominant T^2 part of the low-temperature resistivity (subtracting out the residual resistivity) is due to electron-electron (paramagnon) scattering.

Perhaps the most direct evidence for paramagnon effects will come from tunneling measurements of the reduced density of states. In analogy to the electron-phonon coupling constant λ_p, the effect of λ_s should also be found.[5] We have made tunnel junctions on sputtered films of VN, and although the tunneling results are inconclusive, we have demonstrated the importance and difficulty of obtaining well-ordered films. Disorder can significantly reduce T_c, but its effect on λ_p, λ_s, and μ^* individually is the subject of considerable speculation and is not readily determined. A convenient measure of disorder is the residual resistance ratio, r_R equal to the ratio of the resistances at 300 and 10 K. Our best films have $r_R \sim 2$—5, which is considerably larger than similarly prepared films of the isoelectronic compound NbN. The VN films also display a high degree of polycrystalline epitaxy with the [111] direction normal to the substrate. The temperature-dependent resistivities $\rho(T)$, for two of our samples and one from the literature,[7] are shown to be consistent with the parallel-resistor model,[8] with Matthiesen's rule approximately verified for the ideal resistivity. The maximum resistivity of this model is about 231 $\mu\Omega$ cm and the intrinsic temperature-dependent resistivity at 300 K is about 21 $\mu\Omega$ cm. The coefficient of the T^2 part of the intrinsic low-temperature resistivity is found to be $\approx 9.75\times10^{-4}$ $\mu\Omega$ cm/K^2. These results, along with Josephson tunneling measurements, will be discussed in the following sections.

II. SAMPLE PREPARATION AND CHARACTERIZATION

All films were made reactive dc sputtering using a Plasmax triode source which allowed independent manipulation of the discharge current and target potential (typically 400—700 V). Approximately 25 films were deposited onto polished, single-crystal sapphire substrates with two different orientations, i.e., the sapphire *c* axis parallel (0°) or perpendicular (90°) to the film normal. Optimum substrate temperature for well-ordered films was 600 °C, measured by mounting a Chromel-Alumel thermocouple to the surface of an adjacent substrate on the heater block. The stainless-steel vacuum chamber was pumped sequentially by an oilless rotary pump, liquid-nitrogen-cooled sorption pump, and a closed-cycle helium cryopump which provided a hydrocarbon-free environment. After bakeout the typical residual pressure in the chamber prior to sputtering was 2×10^{-7} Torr *with the heater at* 600 °C. High-purity (99.999%) argon was injected directly into the Plasmax source, whereas the N_2 gas was bled in via one of the chamber ports, both gases being pumped through a liquid-nitrogen-cooled orifice on the cryopump. Typical pressures for the Ar and N_2 were 4 and 1 mTorr respectively, as measured by an ion gauge and a Baratron absolute-pressure manometer. The vanadium target (0.75 in. diam) was prepared by melting VP-grade pellets from MRC in an ultrahigh vacuum (UHV) electron-beam hearth. Because of the sensitivity of T_c to oxygen contamination in transition metals and their compounds, it is important to maintain as low a partial pressure of O_2 as possible. A stringent test of O_2 contamination for any sputtering system is to deposit pure V at moderate rates and check T_c. Our deposition of V films onto unheated substrates at rates of 3 Å/sec resulted in T_c values near 5.0 K, close to the bulk value of 5.3 K. We believe this indicates that there are no significant problems with oxygen contamination in our films. Typical deposition rates of the VN films were 100 Å/min and their thickness was ~4000 Å.

The crystal structure of the deposited films was determined by conventional x-ray diffraction. All films deposited on 600 °C substrates displayed only a single, sharp x-ray line corresponding to reflections from (111) plane of the fcc structure, as shown in Fig. 1. The peak height is about 100 times larger than that found in typical polycrystalline films of VN and NbN, and no other reflections are observed. This indicates that essentially *all* of the grains are growing in the [111] direction. We refer to this as polycrystalline epitaxy, and it is observed for both orientations of the sapphire substrate. The exact role of the substrate in this epitaxial growth has not been investigated, but we believe the dominating factors are the growth conditions rather than compatibility of film and substrate crystal structures. The evidence for this is that NbN (and VN) films sputtered onto single-crystal sapphire by conventional rf and dc diode methods show random orientations,[9] whereas NbN prepared by us under identical conditions as the VN samples always exhibit epitaxial growth in the [111] direction. Since the (111) plane is the one of closest packing for fcc structure, it is not surprising the [111] growth is preferred. Possibly the lower target potentials (400—500 V) compared to conventional magnetron sputtering (1000 V) facilitates the preferred orientation.

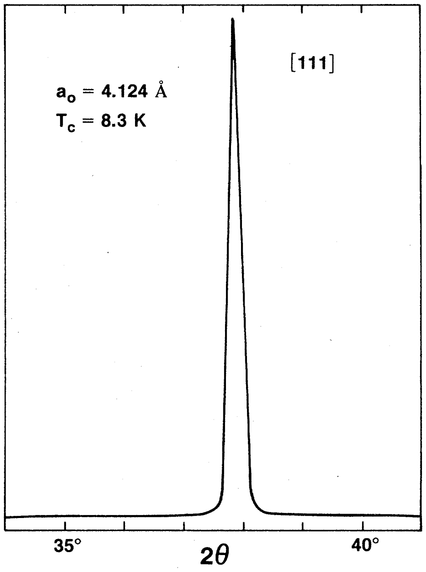

FIG. 1. Typical θ-2θ x-ray diffraction pattern showing the uniaxial growth of the VN films along the [111] direction. The peak height is about 100 times larger than conventional, diode-sputtered films of VN and NbN. No other reflections are observed.

The superconducting transition was determined resistively and in some cases additional inductive measurements were obtained using a SQUID (superconducting quantum interference device) susceptometer. The transition width ΔT_c, determined by the 10%-to-90% criterion, was very small for resistive measurements of the best samples, usually 0.05 K or less. Susceptibility measurements generally displayed values of $\Delta T_c = 0.2$ K or less for the best samples, whereas low T_c films (~4.5 K) had widths as great as 1.0 K. The transition temperature was determined from the midpoint of the transition and the highest value was 8.9 K.

The VN system has been well characterized by a number of experiments on bulk samples.[10,11] Efforts to make thin films by reactive sputtering include the early work of Hofmann-Kraeft[12] and more recently the work of Zhao *et al.*[7] The bulk studies generally agree that the *B*1 fcc phase of VN_x exists in the composition range $0.72 \le x \le 1.00$. The lattice constant increases in a nearly linear fashion from $a_0 = 4.0662$ Å at $x = 0.72$ to $a_0 = 4.1398$ Å at $x = 1.00$. Although the determination

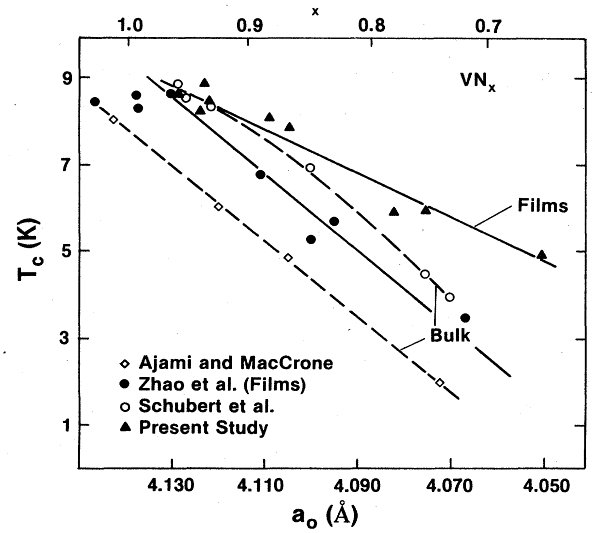

FIG. 2. Superconducting transition temperature T_c vs lattice parameter a_0 for our films as well as bulk and thin-film results from the literature. The data of Ajami and MacCrone (Ref. 10) are representative of other bulk results. The nitrogen content x (top abscissa) corresponds with the values of a_0 obtained from the bulk studies. The VN$_x$ film data point near $a_0=4.050$ Å was not single phase.

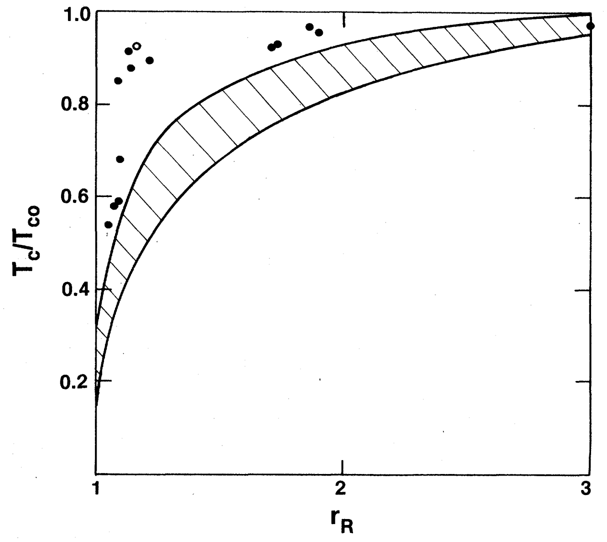

FIG. 3. Superconducting T_c (normalized $T_{c0}=9.25$ K) vs resistance ratio r_R for our VN$_x$ films. Open circle data point is from the work of Zhao *et al.* (Ref. 7). The shaded region corresponds to the universal behavior demonstrated by high-T_c $A15$ compounds as well as the ternary compound ErRh$_4$B$_4$. All the VN$_x$ points lie above the universal curve, clearly demonstrating less sensitivity to disorder.

of nitrogen content in VN$_x$ samples might contain some uncertainty, lattice-constant measurements are quite accurate. Considering this, the dependence of T_c vs a_0 for our films, as well as the bulk[10,11] and thin-film[7] results from the literature, are plotted in Fig. 2. The maximum value of T_c is found near $a_0=4.130$ Å, which corresponds to $x=0.96$ in the bulk studies. The decrease of T_c with decreasing a_0 is consistently found, but the differences between the various sets of data are quite large. For example, at $a_0=4.070$ Å the T_c values range between 2 and 6 K. It might be argued that bulk and thin-film data should not be compared because effects such as strain might change the lattice constant for a given composition in thin films. However, Zhao *et al.*[7] have shown that for a given lattice constant the nitrogen content of thin films is *less* that that of the bulk material,[7] implying that all thin film data should lie below the bulk data in Fig. 2. Clearly this is not the case. Although the lattice constant seems to be consistent indicator of nitrogen content in bulk samples and allows a reasonable estimate in thin films,[7] it is apparent that composition alone does not determine T_c. One possible reason is the amount and type of disorder, and this will be discussed in the next section.

III. RESULTS AND DISCUSSION

A. Disorder

There has been considerable interest in the role of disorder in superconductivity since the pioneering work of Poate *et al.*,[13] who demonstrated the "universal" behavior of T_c vs r_R (the shaded region of Fig. 3) for high-T_c $A15$ compounds. More recently, elemental niobium[14] and the ternary compound[15] ErRh$_4$B$_4$ were shown to follow this universal behavior. Surprisingly, this universal behavior does not depend on the origin or nature of the disorder; thus "as-prepared," radiation-damaged, and ion-implanted samples behave the same. One explanation of the universality of this behavior[16] postulates that it is intrinsic to superconductors whose Fermi energy E_F lies near a sharp peak in the electronic density of states $N(E)$. In this model, disorder shortens the electron lifetimes, thus smearing out $N(E)$ and reducing its value at E_F. This in turn reduces λ_p and hence T_c. Therefore the effect does not depend on how the electron lifetimes (related to the residual resistivity and r_R) are shortened.

The data for our VN films, also shown in Fig. 3, are in clear disagreement with the above universal behavior.[17] Thus the T_c of VN is less sensitive to disorder, and this is consistent with the insensitivity of T_c to radiation damage in experiments[18] on NbN.[19] The explanation may be found in band-structure calculations[20,21] for VN and NbN, supported by experiment,[21] which indicate that E_F lies not to a peak, but midway between a peak and a valley in $N(E)$. Recalling that T_c of low-T_c $A15$ compounds *increases* with disorder because E_F is at a valley in $N(E)$, it is not difficult to imagine a low sensitivity to disorder in VN, as shown in Fig. 3. Calculations[22] of $N(E)$ and E_F for substoichiometric TiN$_x$, which has a similar $N(E)$ at $x=1$ as VN, shows that $N(E)$ is considerably altered, but that $N(E_F)$ changes little (it actually increases slightly), with the increased disorder in going from $x=1$ to $x=0.75$. This increased disorder is not surprising since, for example, at $x=0.75$, the vacancies are about 15 Å apart on average, and should lead to significant electron scattering and concomitant broadening of the structure in $N(E)$.

Based on the changes in $N(E)$ found in Ref. 22 for $x=0.75$, it is surprising that a rigid-band model could be used[10] to describe the T_c variations for bulk VN_x shown in Fig. 2. A simple rigid-band calculation, using $N(E)$ from Ref. 20, assuming each nitrogen atom contributes five electrons to the conduction band, and assuming that $\lambda_p \cong 1.1$ and $\mu^* = 0.13$ for $x=1$, gives $T_c \sim 1.4$ K for $x=0.72$ using the McMillan equation. Although this agrees reasonably well with the bulk data of Ref. 10 shown in Fig. 2, it is clear from the other data shown in Fig. 2 that the composition x (or lattice constant a_0) alone is insufficient to determine T_c. The possibility of oxygen contamination reducing T_c cannot be ruled out. It is also possible, depending on the band structure, that the samples with higher T_c values are actually *more* disordered, due to impurity or grain-boundary scattering. Knowledge of r_R for all samples shown in Fig. 2 would be most helpful to clear us this point.

B. Temperature-dependent resistivity

The temperature dependence of the electrical resistivity $\rho(T)$ was measured on three films using standard four-terminal techniques (see Fig. 4). The behavior of $\rho(T)$ is similar to the high-T_c $A15$ compounds, i.e., an initial T^2 dependence at low temperatures followed by a region of negative curvature.[23] Including the VN film reported in Ref. 7, the coefficient of the T^2 term is found to decrease monotonically with the measured residual resistivity. Such behavior was also found by Gurvitch *et al.*[8] for Nb_3Sn. A parallel-resistor model has been proposed[24] to explain the saturation of $\rho(T)$. It assumes a temperature- and sample-independent resistivity ρ_m in parallel with the sum of the ideal residual (ρ_0) and ideal temperature-dependent resistivities. To check this model at low temperatures, we calculate the parallel combination of ρ_m and $\rho_0 + AT^2$ to be

$$\rho = \rho_0' + A'T^2 - B'T^4 ,$$

where $\rho_0' = \rho_m \rho_0/(\rho_m + \rho_0)$, $A' = \rho_m^2 A/(\rho_m + \rho_0)^2$, and $B' = \rho_m A^2/(\rho_m + \rho_0)^2$ are the coefficients determined by fitting the experimental data. Neglecting B' for the moment, we calculate ρ_m as a function of A for each of the samples listed in Table I, using (from above)

$$\rho_m = \rho_0'/(1 - \sqrt{A'/A})$$

as a function of A. When $A = 9.75 \times 10^{-4}\ \mu\Omega\,\text{cm/K}^2$, the values of ρ_m are sufficiently close (from Table I, $231 \pm 1\ \mu\Omega\,\text{cm}$) to demonstrate the consistency of the model. The temperature at which the B' term equals the A' term is $(\rho_m/A)^{1/2} \sim 400$ K, independent of sample, which justifies its neglect below 40 K where the T^2 term is found. The implication of this agreement is that the T^2 term does not depend on residual resistivity. The value of ρ_m is not too far from the maximum metallic resistivity of 251 $\mu\Omega$ cm, calculated by Hake[25] for NbN. Changing only the lattice constant to the smaller value for VN, Hakes's analysis yields a maximum metallic resistivity of about 236 $\mu\Omega$ cm for VN. One also finds $\rho_0 = \rho_0'\sqrt{A/A'}$, and ρ_T, which is the difference in the ideal resistivity at 10 K and 300 K, is given in terms of the measured resistivity $\rho_{300\text{ K}}'$, by

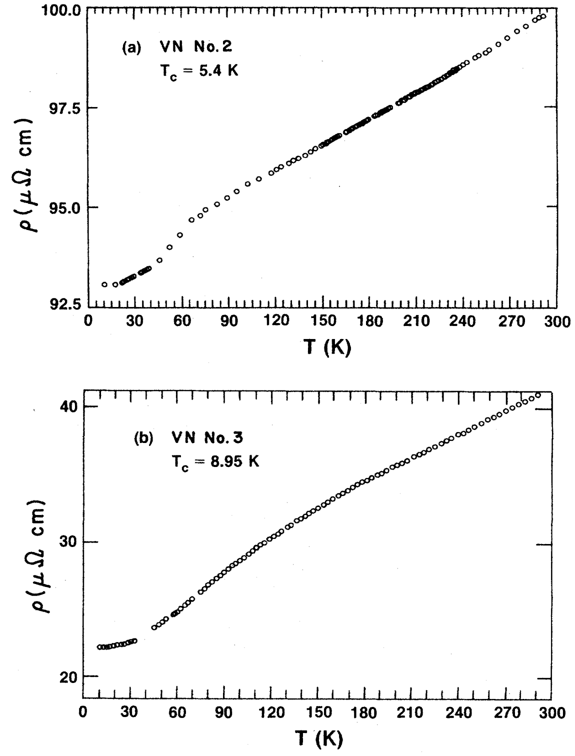

FIG. 4. Resistivity versus temperature for two VN films with resistance ratios (a) $r_R = 1.07$ for sample 2 and (b) $r_R = 1.86$ for sample 3.

TABLE I. Parallel-resistor model. Parameters were determined using the parallel-resistor model for our samples shown in Fig. 4 and that of Fig. 3 of Ref. 7. The resistivities of Ref. 7 were reduced uniformly by 3% to significantly improve the fit. This is justified since film thicknesses can rarely be measured with such accuracy. The parameter A is $9.75 \times 10^{-4}\ \mu\Omega\,\text{cm/K}^2$.

Sample	ρ_0' ($\mu\Omega$ cm)	A' ($10^{-4}\ \mu\Omega$ cm/K^2)	ρ_m ($\mu\Omega$ cm)	ρ_0 ($\mu\Omega$ cm)	ρ_{300} ($\mu\Omega$ cm)	ρ_T ($\mu\Omega$ cm)
3	22	7.9	230	25	42	26
Ref. 7	55	5.6	232	73	65	17
2	93	3.5	232	154	100	21

$$\rho_T = \frac{(\rho'_{300\ \mathrm{K}} - \rho_0)\rho_m + \rho'_{300\ \mathrm{K}}\rho_0}{\rho_m - \rho'_{300\ \mathrm{K}}} \ .$$

The agreement for the three samples is much poorer than for fitting AT^2 (last column of Table I). Forcing a minimum deviation in ρ_T for the three samples results in a lower ρ_m ($\sim 190\ \mu\Omega\,\mathrm{cm}$) and a larger variation in the T^2 term, so a more complete study is desirable to ascertain whether Matthiesen's rule is followed for the ideal resistivity.

The above discussion casts little light on the origin of the T^2 term in VN, which has been suggested[7] as being due to electron-electron (paramagnon) scattering. However, it seems unlikely that the very similar results[8] found in $\mathrm{Nb_3Sn}$ would have different origins (paramagnon effects are expected to be small in $\mathrm{Nb_3Sn}$). Also, a stronger electron scattering due to paramagnons compared to phonons would imply a large coupling constant λ_s, making it unlikely that VN would be superconducting at 9 K (see comments in the Introduction). A more reasonable picture is that VN behaves like pure V, i.e., $\lambda_p > \lambda_s$, but nevertheless T_c is reduced by spin fluctuations.[6] Available experimental data on VN, including our results, are consistent with the theoretical values ($\lambda_p \sim 1.1$ and $\lambda_s \sim 0.3$) suggested by Rietschel *et al.*[2] in their "intermediary" situation in which T_c would be ~ 22 K if spin fluctuations are ignored. The similarity of the $\rho(T)$ behavior in VN and other high-T_c superconductors is understandable within this picture.

C. Tunneling

All of the better films were either used for resistivity measurements or lost in susceptibility measurements. Several films with T_c near 7 K were used to test the method of plasma oxidation of the VN surface to form tunnel barriers. Even though vanadium oxides had been previously found unsuitable for barriers, this method was tried since it had been successful in fabricating low-leakage NbN junctions.[26] In Fig. 5 conductance data for such a junction using a Pb counterelectrode are shown. A well-defined sum gap is observed, leading to a value of $\Delta = 1.09$ mV for this VN sample. The ratio $2\Delta(0)/k_B T_c$ is 3.78, somewhat reduced from the value 3.99 for stoichiometric VN.[7] The change from weak-coupling to strong-coupling behavior as T_c approaches its maximum value is similar to that observed[27] in NbN as well as in *A*15's. Leakage conductance for this junction is relatively high (~ 25 %), and a more extensive investigation of the plasma-oxidation technique using higher-T_c VN films is currently underway. Attempts to restore the surface of films, previously used for resistivity, by mild ion bombardment, followed by plasma oxidation, were unsuccessful. Good junctions were formed but superconductivity in the surface layer of the VN films was severely degradated.

D. Josephson tunneling

Josephson tunnel junctions were fabricated on some VN films using light-sensitive semiconducting CdS films for tunneling barriers and indium counterelectrodes. The basic fabrication technique and the behavior of Josephson tunnel junctions realized by CdS barrier depositions on different base electrodes is described in Ref. 28 and 29. The VN-CdS-In junctions made by this technique displayed *I*-*V* curves with well-defined sum-gap structures, but with leakage currents of the order of 30%–40%. The magnetic field dependence of the zero-voltage Josephson current, in "dark" conditions, is reported in Fig. 6 for one of our samples with $T_c(\mathrm{VN}) = 8.6$ K. The well-defined Fraunhofer-type behavior indicates good barrier uniformity. Using the magnetic field periodicity observed in the diffraction pattern ($\Delta B_{\min} \cong 0.35$ G), the film thickness ($t_{\mathrm{VN}} = 500$ nm, $t_{\mathrm{CdS}} = 60$ nm, $t_{\mathrm{In}} = 150$ nm), the junction length perpendicular to the field (233 μm), and the bath temperature (3.33 K), a value of the London penetration depth of VN equal to 130 ± 10 nm can be deduced.[28] This value is consistent with the low resistivity of our VN films compared to NbN films, in which a value of 500 nm is found. The temperature dependence of the zero-field Josephson current was also measured for these samples and the behavior was found to

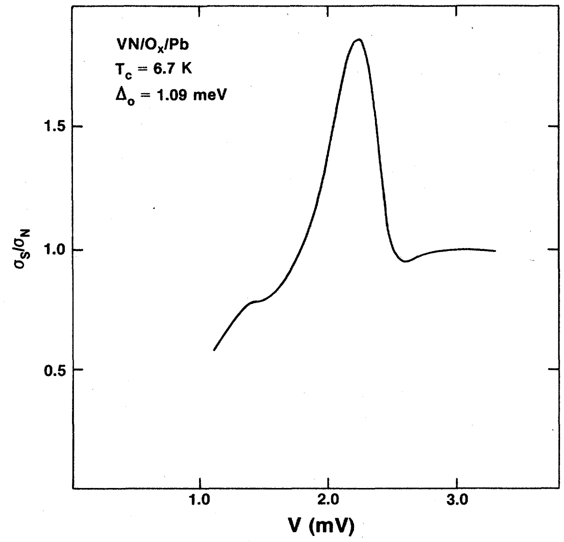

FIG. 5. Normalized dynamic conductance versus bias for VN/O_x/Pb tunnel junction fabricated by plasma oxidation of the VN surface. A well-defined sum gap is observed leading to the ratio $2\Delta(0)/k_B T_c = 3.78$.

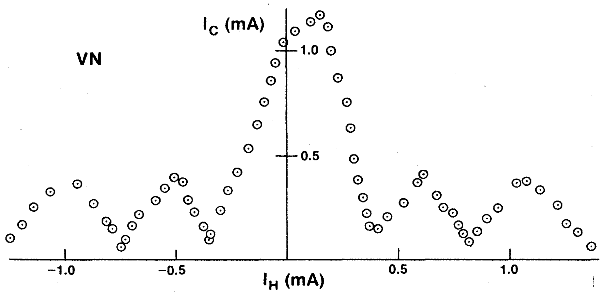

FIG. 6. Maximum Josephson current versus magnet current for a VN tunnel junction utilizing a CdS artificial barrier.

be consistent with the proximity effect model that generally applies to CdS-barrier Josephson junctions.[29]

IV. SUMMARY

We have reported on high-quality VN_x films made by dc triode sputtering. The films exhibit a very high degree of preferential orientation with the [111] direction normal to the sapphire substrate. Measured T_c are higher than bulk for the same lattice constant, especially for N deficient samples. The role of disorder in reducing T_c is found to be much less than for high-T_c $A15$ superconductors. A T^2 term in the temperature-dependent resistivity is found, as in the case of Nb_3Sn, and a parallel-resistor model successfully accounts for differences amongst samples with a maximum resistivity of 231 $\mu\Omega$ cm. Quasiparticle tunneling confirms that VN_x is strong coupling, while the Frauenhofer pattern observed in Josephson tunneling through a CdS barrier reveals the penetration depth to be 1300±100 Å.

Note added. Recent resistivity measurements on our cleanest sample ($r_R \sim 4$) show $\rho(T) \propto T^3$ for $T_c < T < 30$ K rather than the T^2 dependence for the lower r_R samples shown in Fig. 4. This crossover from T^2 to T^3 as resistivity is reduced is in agreement with the empirical trends noted by Gurvitch.[30]

ACKNOWLEDGMENTS

We thank M. Gurvitch for helpful discussions. This work is supported in part by the U. S. Department of Energy. One of us (J.Z.) would like to acknowledge the receipt of a Cottrell grant from the Research Corporation (New York, NY).

---

[1] L. E. Toth, *The Transition Metal Carbides and Nitrides* (Academic, New York, 1971).

[2] H. Rietschel, H. Winter, and W. Reichardt, Phys. Rev. B **22**, 4284 (1980).

[3] H. Rietschel and H. Winter, Phys. Rev. Lett. **43**, 1256 (1979).

[4] D. M. Burnell, John Zasadzinski, R. J. Noer, E. L. Wolf, and G. B. Arnold, Solid State Commun, **41**, 637 (1982).

[5] J. M. Daams, B. Mitrović, and J. P. Carbotte, Phys. Rev. Lett. **46**, 65 (1981).

[6] T. W. E. Tsang, K. A. Gschneidner, F. A. Schmidt, and D. K. Thome, Phys. Rev. B **31**, 235 (1985).

[7] B. R. Zhao, L. Chen, H. L. Luo, M. D. Jack, and D. P. Mullin, Phys. Rev. B **29**, 6198 (1984).

[8] M. Gurvitch, A. K. Ghosh, H. Lutz, and M. Strongin, Phys. Rev. B **22**, 128 (1980).

[9] A. M. Cuculo, L. Maritato, A. Saggese, and R. Vaglio, Cryogenics **24**, 45 (1984).

[10] F. I. Ajami and R. K. MacCrone, J. Phys. Chem Solids **36**, 7 (1975).

[11] W. K. Schubert, R. N. Shelton, and E. L. Wolf, Phys. Rev. B **23**, 5097 (1981).

[12] B. Hofmann-Kraeft, Ph.D. thesis, Kernforschungszentrum und Universität Karlsruhe, Karlsruhe (1977).

[13] J. M. Poate, L. R. Testardi, A. R. Storm, and W. M. Augustyniak, Phys. Rev. Lett. **35**, 1290 (1975).

[14] C. Camerlingo, P. Scardi, C. Tosello, and R. Vaglio, Phys. Rev. B **31**, 3121 (1985).

[15] J. M. Rowell, R. C. Dynes, and P. H. Schmidt, Solid State Commun. **30**, 191 (1979); K. E. Gray, J. Zasadzinski, R. Vaglio, and D. G. Hinks, Phys. Rev. B **27**, 4161 (1983).

[16] L. R. Testardi and L. F. Matthiess, Phys. Rev. Lett. **41**, 1612 (1978).

[17] There is also clear disagreement with the "universal behavior" in $SmRh_4B_4$, but this is associated with disorder enhanced spin flip scattering from the magnetic ions of Sm. See B. D. Terris, K. E. Gray, R. T. Kampwirth, J. Zasadzinski, and R. Vaglio, Phys. Rev. B **30**, 5370 (1984).

[18] D. Dew-Hughes and R. Jones, Appl. Phys. Lett. **36**, 856 (1980).

[19] For NbN, correlations with r_R are difficult because of the large grain-to-grain resistances leading to $r_R < 1$ in some cases. See, e.g., R. T. Kampwirth and K. E. Gray, IEEE Trans. Magn. **MAG—17**, 565 (1981); R. T. Kampwirth, D. W. Capone II, K. E. Gray, and A. Vincens, IEEE Trans. Magn. **MAG—21**, 459 (1985).

[20] M. Dacorogna, T. Jarlborg, A. Junod, M. Pelizzone, and M. Peter, J. Low Temp. Phys. **57**, 629 (1984).

[21] J. Pflüger, J. Fink, W. Weber, K.-P. Bonhen, and G. Crecelins, Phys. Rev. B **30**, 1155 (1984).

[22] J. Klima, G. Schadler, P. Weinberger, and A. Neckel, J. Phys. F **15**, 1307 (1985).

[23] G. W. Webb, Z. Fisk, J. J. Engelhardt, and S. D. Bader, Phys. Rev. B **15**, 2624 (1977).

[24] H. Wiesmann, M. Gurvitch, H. Lutz, A. K. Ghosh, B. Schwartz, M. Strongin, P. B. Allen, and J. W. Halley, Phys. Rev. Lett. **38**, 782 (1977).

[25] R. R. Hake, Appl. Phys. Lett. **10**, 189 (1967).

[26] J. Zasadzinski, W. Gai, R. Vaglio, A. Saggese, and L. Maritato (unpublished).

[27] K. E. Gray, R. T. Kampwirth, D. W. Capone II, and R. Vaglio (unpublished).

[28] A. Barone and G. Paterno, *Physics and Applications of the Josephson Effect* (Wiley, New York, 1982).

[29] F. Andreozzi, A. Barone, M. Russo, G. Paterno, and R. Vaglio, Phys. Rev. B **18**, 6035 (1978).

[30] M. Gurvitch, Bull. Am. Phys. Soc. **30**, 633 (1985) and unpublished.