Channeling in V_3Si: Atomic Displacements and Electron-Phonon/Defect Interactions

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^{4}He channeling measurements of minimum yields in single-crystal V_3Si have been compared with those calculated with the Karlsruhe phonon density of states. Atomic displacements versus temperature and defect level have been so obtained. *In situ* radiation-damaged crystals show $\sim 10^{-1}$-Å displacements and significant phonon stiffening. The observations explain the large changes in electrical resistivity in the defect state, and suggest that both thermal and static (defect) displacements diminish the electron-phonon/defect interactions, and to equivalent degrees.

We report the results of a ^{4}He channeling study of the dynamic and static atomic displacements of V in the A-15-structure compound V_3Si. The measured values of the channeling minimum yield χ_{min} are found to be appreciably smaller than expected from elemental crystals. This probably results from the reduced symmetry about the atomic rows in the V_3Si lattice. Measurements of χ_{min} further show that ^{4}He radiation damage, which causes large reductions in the superconducting T_c, appears to involve static displacements of atoms from lattice sites and not antisite defects alone. The defects also induce considerable phonon stiffening. These effects can be correlated with the large changes in electrical resistivity of damaged V_3Si films, and suggest that both thermal and static (defect) displacements reduce the electron-phonon/defect interactions, and many of the anomalous properties, in qualitatively similar ways.

Channeling and backscattering studies were performed using 1.9-MeV ^{4}He particles in the ⟨100⟩ and ⟨110⟩ directions for the V atoms in four samples of V_3Si.[1] The samples were plate shaped [two each with (100) and (011) orientations] and cut from a single crystal whose T_c was ~17 K, electrical-resistivity ratio $\rho(300\text{ K})/\rho(20\text{ K}) \sim 22$, and which exhibited (from ultrasonic studies) some evidence of the Batterman-Barrett transformation[2] beginning at 21 K.

Measurements were made from 20 to 300 K for a ⟨100⟩ direction, and from 20 to 670 K for a ⟨110⟩ direction. *In situ* radiation-damage studies were also carried out with use of higher ^{4}He beam currents in a random direction. Care was taken to establish that the beam currents used in the channeling studies did not cause significant damage in this defect-prone compound.

Figure 1 shows $\chi_{min}^{expt}(T)$ for the V atoms in the ⟨100⟩ and ⟨110⟩ directions of V_3Si. About ten separate determinations of χ_{min} for each direction were made at 300 K on the four samples, and these values agreed to within ~± 7%, which is approximately the statistical counting error. The temperature dependence was measured once for the ⟨110⟩ direction and eight times for the ⟨100⟩ direction.

To have an expression for χ_{min}, we have generalized the results for elemental materials[4] and obtain

$$\chi_{min}^{theor} = (\pi/A)\textstyle\sum_i \nu_i C_i \tau_i^2 (1+\zeta_i^{-2})^{1/2}, \quad (1)$$

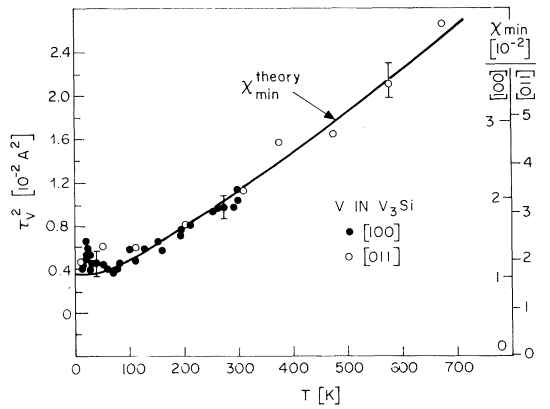

FIG. 1. Observed minimum yield χ_{min} from channeling and the derived two-dimensional mean-square vanadium displacements τ_V^2 vs temperature for ⟨100⟩ and ⟨110⟩ directions in V_3Si. Several representative error bars are indicated. Also shown are χ_{min}^{theor} for ⟨100⟩ and ⟨110⟩. The left-hand scale for τ_V^2 is slightly nonlinear and offset to correct for the oxide-layer contribution (Ref. 3).

where A is the area of a surface unit cell, ν_i is the number of rows in the surface cell made up of the ith (i is either V or Si) constituent, and τ_i^2 is the two-dimensional mean-square displacement of the ith constituent perpendicular to the channeling direction. The C_i's are discussed below. $\tau^2 = \overline{u_{11}^2} + \overline{u_{22}^2}$ (u's are the one-dimensional Debye-Waller displacements), and is composed of static, zero-point, and thermal-vibrational contributions. ζ^{-2} is defined in Ref. 4 and is of order $\lesssim 0.5$. Equation (1) holds for an axial channeling direction in which all rows are made up of one constituent only. In V_3Si, this condition applies for the $\langle 100\rangle$ and $\langle 110\rangle$ directions ($\nu_V = 5$ and 6, respectively; $\nu_{Si} = 2$ and 2, respectively).

For χ_{min}^{theor} we have calculated τ_V^2 and τ_{Si}^2 from the phonon densities of states $G(\omega)$ at 4 and 300 K, recently determined by Schweiss and coworkers.[5] In this calculation[6] we have assumed an isotropic τ_V^2 and a decomposition of $G(\omega)$ into $g_V(\omega)$ and $g_{Si}(\omega)$ in such a way that for $\omega \to 0$ the V vibrations dominate (in-phase motion of V and Si) and for $\omega \to \omega_{max}$ the Si vibrations dominate (out-of-phase motion of V and Si). While the values for τ_{Si}^2 depend strongly on the choice of the $G(\omega)$ decomposition, τ_V^2 and χ_{min}^{theor} and rather insensitive to it. When we adjust the decomposition such that τ_{Si}^2(300 K) agrees with recent x-ray data,[7] τ_V^2 also agrees very well. Very recent measurements by Staudenmann of τ_V^2 and τ_{Si}^2 at 13 K also agree with our calculations. We note that these x-ray results reasonably justify the assumption of isotropy for τ_V^2.

Values of χ_{min} obtained from Eq. (1) using the calculated τ's[3] are much larger than the experimental values if $C_V = 3.0$ and $C_{Si} = 1.7$, as suggested for elemental materials.[4] However, the C's represent enhancement over ergodic behavior caused in large part by focusing effects of the lattice on the ion trajectories.[8] In V_3Si, the V and Si rows are intermingled and have low symmetry so that the focusing effects are reduced. Hence the C's were varied to obtain the best agreement with experiment. Because χ_{min}^{theor} is relatively insensitive to C_{Si}, C_{Si}/C_V was kept fixed at 1.7/3.0. The best fit was obtained with $C_V = 1.7$ and $C_{Si} = 1.0$ with results as shown by the theoretical curve in Fig. 1. The fit is, at all temperatures and for both axes, within the experimental uncertainty. If small C values are characteristic of compounds, they should be observable in other cases.

The great susceptibility of A-15 superconductors to radiation damage[9] was studied by *in situ* damage in a random direction at room temperature followed by channeling measurements. From the previous analysis we are now able to obtain values of τ_V^2 for the damaged crystal from the channeling data. Figure 2 shows the observed χ_{min} and the deduced τ_V^2 from 20 to 300 K for $\langle 100\rangle$ channeling after damage doses of 0.75, 1.5, and 2.5×10^{17} cm^{-2} of 1.9-MeV ^{4}He particles. Based on sputtered-film measurements these doses will reduce T_c to ~ 8, 5, and 2 K (the low T_c-saturation value of Poate *et al.*[9]), respectively (see Fig. 3). We find that for $\langle 100\rangle$ (and, from limited testing, $\langle 110\rangle$) channeling, these doses significantly increase χ_{min}. This behavior is not typical for a metal where the induced defects largely anneal out at 300 K, but is more characteristic of a covalently bonded semiconductor for which such defects lead to a metastable quasi-amorphous state.[10]

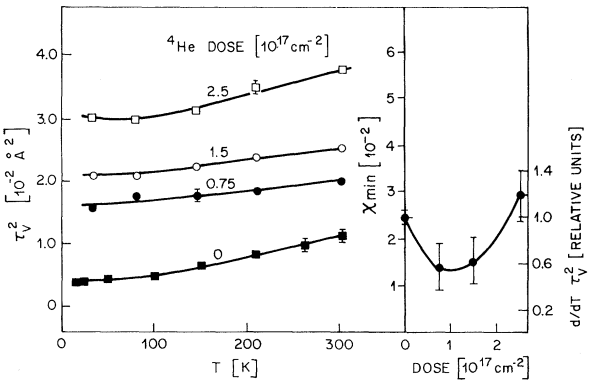

FIG. 2. Observed minimum yield and derived two-dimensional τ_V^2 for $\langle 100\rangle$ in V_3Si with and without damage as a function of temperature. The inset shows $(d/dT)\tau_V^2$ at 300 K.

Simple antisite disorder, while retaining a perfect lattice, will not produce measurable increases in χ_{min}. Our results suggest either 1–2% localized regions of complete disorder or small bond distortions for most of the atoms producing displacements, τ_V, from the atomic rows. Previous x-ray studies,[11] however, have indicated small bond distortions throughout the bulk of the sample. Calculations of the Debye-Waller factor for the observed τ^2 predict an observable decrease (~ 50% for Cu $K\alpha$ and $\theta > 45°$) in the intensity of high-angle x-ray diffraction lines in the highly damaged state—a result qualitatively consistent with observation.[11] We have recently learned of channeling and radiation-damage experiments carried out at 300 K in V_3Si by Meyer and Seeber[12] who also conclude the existence of the V

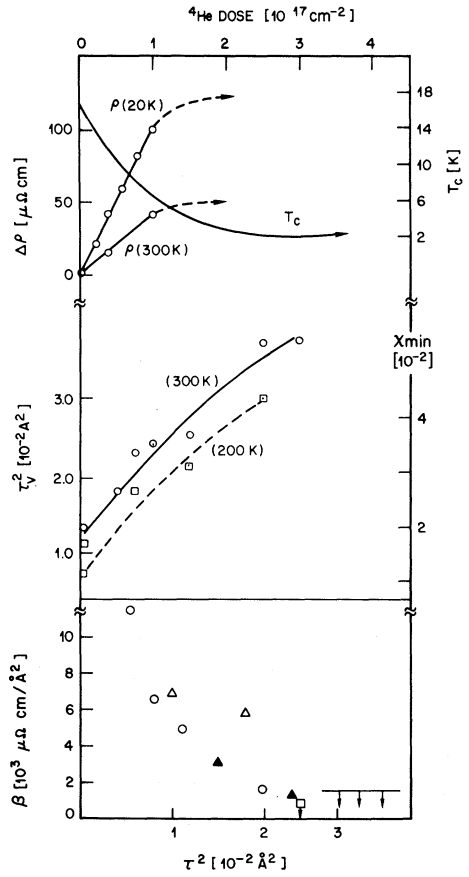

FIG. 3. Change in T_c, electrical resistivity ρ, and τ_V^2 as a function of ^{4}He dose. Botton panel shows universal decrease of β vs τ^2. ○, β_{th} (undamaged); □, β_{th} (damaged); △, β_d (20 K); ▲, β_d (300 K).

displacements.

Figure 2 (inset) shows, in addition, that with increasing damage τ_V^2 has a minimum in the thermal contribution τ_{th}^2 of about (60 ± 20)% of the ideal-crystal value when T_c has been reduced to 5 K. Since $\tau_{th}^2 \propto \langle \omega_{phon}^{-2} \rangle T$ (for T greater than $\frac{1}{5}$ of the Debye temperature Θ_D), the decrease of τ_{th}^2 indicates a significant stiffening of the phonons with defects. This observation is consistent with recent sound-velocity[13] and specific-heat[14] measurements on neutron-damaged V_3Si. Phonon stiffening with increasing temperature has been previously reported.[2,5]

Figure 3 shows the increase in $\langle 100 \rangle \tau_V^2$ with increasing dose for samples damaged at 300 K and analyzed at 20 and at 300 K. Also shown are the increases in electrical resistivities ρ at 20 and 300 K and the decrease in T_c with dose as observed[15,9] in sputtered films which have properties similar to the bulk crystal. Note that χ_{min} continually increases with dose beyond the point where T_c and ρ saturate. (The saturation trend in ρ is from a new datum point at high dose.)

Figure 3 further shows that the large increases in ρ with ^{4}He dose are in contradiction to Matthiessen's rule.[15] We may analyze this behavior by writing

$$\Delta\rho = \beta_{th}\Delta\tau_{th} + \beta_d\Delta\tau_d = \Delta\rho_{th} + \Delta\rho_d. \quad (2)$$

Here β_{th} represents the electron-phonon coupling [proportional to McMillan's $N(0)J^2$] and β_d is the electron-defect coupling. τ_{th}^2 and τ_d^2 are the thermal and static (defect) mean-square displacements of the V atoms. Equation (2) should be approximately valid for $T \gg \Theta_D/5$ with regard to $\Delta\rho_{th}$, and at all T with regard to $\Delta\rho_d$.

From Figs. 2 and 3, and Eq. (2), we estimate that at low doses $\beta_d(20\text{ K}) \sim 7\times10^3$ μΩ cm/Å^2 and $\beta_d(300\text{ K}) \sim 3.3\times10^3$ μΩ cm/Å^2. At high doses ($>1.5\times10^{17}$ cm^{-2} ^{4}He) the increase in electrical resistivity with dose tends to saturate[15] while τ_d^2 continues to increase. Thus β_d is considerably reduced for large τ_d^2.

For undamaged V_3Si we find $\beta_{th}(300\text{ K}) \sim 5\times10^3$ μΩ cm/Å^2 from the ratio of $\Delta\rho/\Delta T$ (Ref. 15) and $\Delta\tau^2/\Delta T$ (Fig. 1). However, at 500 K we obtain $\beta \sim 2\times10^3$ μΩ cm/Å^2.

For elemental[16] V ($T_c \approx 5$ K), $\beta_{th} = 2\times10^3$ and $\beta_d = 2.5\times10^3$ μΩ cm/Å^2 at 300 K. It can thus be concluded that the large increase in ρ for V_3Si induced by radiation damage results not only from the atypically large τ_d^2 but also from the large electron-defect coupling as well.

Furthermore, the failure of Matthiessen's rule in V_3Si appears to be a consequence of the fact that β_{th} (like β_d) is reduced with increasing τ^2. For example, samples damaged with 10^{17} cm^{-2} ^{4}He ($T_c \sim 6$ K) show $\tau_{th}^2(300\text{ K})$ reduced by ~ 40% (Fig. 2), but $\rho_{th}(300\text{ K})$ reduced by ~ 70% (Fig. 9 of Ref. 15). More generally, Fig. 3 (bottom panel) displays values of β obtained from both thermal and defect contributions to resistivity as a function of τ^2 over a wide range in temperature and radiation dose. The reduction of β with τ^2 is similar for both cases and shows that the anomalous temperature dependence of ρ is consistent with the defect dependence when comparison is made at the same values of τ^2. In addition, since the electron-phonon/defect parameter β is largest when the atoms are closest to the (unstable) lattice sites, both temperature and defects may stabilize the structure.

This universal reduction of the electron-phonon/defect interactions with increasing τ^2 may apply

to other properties such as T_c, the sound velocity, and magnetic susceptibility. The softening of the phonon frequencies due to the electron-phonon interactions, for example, is proportional[17] to β and therefore will be comparably smaller for increased thermal and defect τ^2. In general, we expect the behavior at lower temperatures in the defect state will be qualitatively similar to that at higher temperatures (i.e., with equivalent τ^2's) for the defect-free state.

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[1]The channeling measurements described here sample approximately the first 300 Å below the surface of the crystal with a 0.5-mm-diam beam.

[2]For further details see L. R. Testardi, Rev. Mod. Phys. 47, 637 (1975).

[3]In this calculation we have included surface effects due to a 60-Å oxide layer, which was found to yield $\chi_{\min}^{ox} = 0.003$, 0.004, and 0.005 for 20, 300, and 500 K, respectively, along [001].

[4]J. H. Barrett, F. Fujimoto, K. Komaki, and Y. Hashimoto, Rad. Effects 28, 119 (1976). See also, J. H. Barrett, Phys. Rev. B 3, 1527 (1971).

[5]P. Schweiss, in Technistitut Festkörperphysik, Gesellschaft für Kernforschung Karlsruhe, Progress Report No. KFK 2054, 1974 (unpublished), p. 12; E. Schneider, P. Schweiss, and W. Reichardt, in *Superconductivity in d- and f-Band Metals*, edited by D. M. Douglass (Plenum, New York, 1976).

[6]A. Maradudin, E. Montroll, G. Weiss, and I. Ipatova, in *Lattice Dynamics in the Harmonic Approximation* (Academic, New York, 1971), 2nd ed., p. 303.

[7]R. Flükiger, J.-L. Staudenmann, and P. Fischer, J. Less-Common Met. 50, 253 (1976).

[8]J. H. Barrett, Phys. Rev. Lett. 31, 1542 (1973).

[9]J. M. Poate, R. C. Dynes, L. R. Testardi, and R. H. Hammond, Phys. Rev. Lett. 37, 1308 (1976), and references therein.

[10]For additional references and discussion see L. C. Kimerling and J. M. Poate, in *Particles and Fields—1974*, AIP Conference Proceedings No. 23, edited by C. E. Carlson (American Insitute of Physics, New York, 1975), Chap. 1, p. 126.

[11]R. C. Dynes, J. M. Poate, L. R. Testardi, A. R. Storm, and R. H. Hammond, IEEE Trans. Magn. 13, 640 (1977); J. M. Poate, L. R. Testardi, A. R. Storm, and W. M. Augustyniak, Phys. Rev. Lett. 35, 1290 (1975).

[12]O. Meyer and B. Seeber, to be published.

[13]M. Sarachik, F. Smith, A. Guha, and L. R. Testardi, to be published.

[14]R. Viswanathan, R. Caton, and D. Cox, to be published.

[15]L. R. Testardi, J. M. Poate, and H. J. Levinstein, Phys. Rev. B 15, 2570 (1977).

[16]W. B. McIntire and J. B. Cohen, Acta Metall. 23, 953 (1975). A factor-of-2 difference is noted between our two-dimensional τ^2 and the one-dimensional x-ray μ_S^2 of this reference.

[17]C. M. Varma and W. Weber, to be published.