Effects of Disorder on the Transition Temperature and Transport Properties of a Low-T_c $A15$ Superconductor: Mo_3Ge

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In Mo_3Ge, an $A15$ superconductor with a low transition temperature of 1.45 K, T_c is found to increase with increasing disorder. We also find that the density of states increases as T_c increases. The transport properties indicate an "ideal" T^5 behavior with Matthiessen's rule being approximately obeyed, for small amounts of damage. For larger amounts of damage, saturation is found in the transport properties even though λ, the electron-phonon coupling, increases as is evidenced by the increasing T_c.

The mechanism for the reduction of T_c in high-T_c $A15$ superconductors has remained a source of some controversy. Various explanations dwell on a sharp density of states in a one-dimensional -type structure which is easily decreased by disorder.[1] Some explanations have been based on gap anisotropy,[2] another point of view has focused on the nature of the defect which causes the disorder,[3] and finally some arguments have been based on general smearing of the density of states with disorder.[4,5]

In this paper we report and analyze a number experiments on Mo_3Ge,[6] where T_c increases with α-particle damage, and we suggest a general picture regarding the whole family of $A15$ superconductors.

Mo_3Ge samples were α-particle irradiated at room temperature in the Brookhaven National Laboratory tandem Van de Graaff accelerator. At each successive radiation, the T_c and ρ_0 of the samples were measured.

The general results showing that T_c increases with ρ_0 are shown in Fig. 1. Two samples were measured, one made by sputtering from a Mo_3Ge ingot, and one sample was made by e-beam evaporation onto a heated substrate. Both samples are seen to give similar results. The density of states has also been determined by measuring $(dH_{c2}/dT)_{T_c}$ and ρ_0 in a manner previously described[4] and, although there is significant scatter in the data, it is found that $n(E_F)$ (density of states at the Fermi energy) increases about 50% with increasing ρ_0.

An important question concerns whether the observed effects are in the Mo_3Ge or whether the increased T_c's are due to some free Mo whose T_c goes up with irradiation. We think that this latter case is improbable for a variety of reasons. (a) X-ray diffraction shows no second phase. (b) Microprobe analysis shows that if anything the samples described here were somewhat Ge rich. (c) The T_c of Mo does not increase significantly when it is irradiated with rare gas particles.[7] Furthermore, in order to test whether Mo in the presence of Ge can be disordered by radiation damage, two experiments were tried. A sample of Mo containing a few percent of Ge and also a ~1000-Å Mo film covered with Ge were both α irradiated to a dose where large T_c changes (~1 K) were already observed in Mo_3Ge. T_c was below 1.2 K in both samples after this dose, showing the effects of irradiation are small even in "dirty" Mo, and furthermore the fact that the thin Ge covered film also had a transition below 1.2 K shows that disordered Mo is not formed by

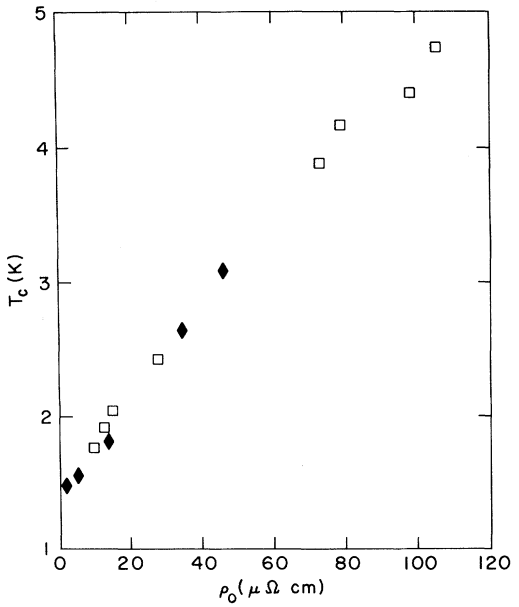

FIG. 1. T_c vs ρ_0 for two Mo_3Ge samples. Squares, sputtered at room temperature and annealed at 750°C; diamonds, electron-beam evaporated onto hot substrate (800°C).

radiation-enhanced diffusion of Ge into the Mo film. (d) Critical-current measurements have been made, and when edge effects are accounted for, the calculated current densities are of the order of 10^7 A/cm^2, which is the value expected for the depairing current. Hence the data are consistent with most of the sample volume being superconducting.

It is a common feature of high-T_c $A15$ compounds that T_c decreases as $n(E_F)$ goes down because of disorder.[4] We find the same correlation between T_c and $n(E_F)$ but now disorder has the opposite effect on $n(E_F)$; it increases with increasing ρ_0. We have estimated $n(E_F)$ by measuring $(dH_{c2}/dT)_{T_c}$ and ρ_0, as previously done for the high-T_c $A15$'s,[4] and we find that $n(E_F)$ changes about a factor of about 1.5 as T_c changes to 5 K, which is enough to explain these T_c changes. However, the precision of the density-of-states measurement is not adequate to say that the T_c changes are totally explained by increases in $n(E_F)$. Actually, a 50% increase in $n(E_F)$ is somewhat higher than the 25% change in λ necessary to account for the T_c changes. Besides the errors in the measurement itself, some of this difference could be accounted for by changes in I^2, the electron-phonon matrix element, which is expected to decrease[8] with increasing lattice parameter. We measure about a 1% increase in lattice parameter at $\rho_0 \sim 80$ $\mu\Omega$ cm, which could mean a 5% to 10% change in I^2. In the clean case we find that $n(E_F) \sim 0.33$ states/eV atom in Mo_3Ge compared to ~1 state/eV atom for Nb_3Sn.

A simple explanation of this somewhat surprising behavior of $n(E_F)$ may be given as follows: One of the characteristic features of the $A15$ compounds containing transition metals and s-p elements (Sn, Al, Ge) is a d band which consists of a dense set of bonding bands at low energies and a similar set of antibonding bands at higher energies. The corresponding two peaks (highly structured) in the density of states are separated by a region of low density of states. This situation is extremely analogous to that which prevails in the bcc transition metals. Thus, for a compound with the d band something like $\frac{1}{3}$ or $\frac{2}{3}$ filled one expects a large $n(E_F)$, while for a half-filled d band $n(E_F)$ is expected to be small. An example of the former case would be Nb_3Sn and an example of the latter is Mo_3Ge. Imagine now that the metals get very disordered. It is reasonable to expect that now the bonding and antibonding bands are no longer split, and that the density of states has a peak in the middle. Thus $n(E_F)$ for a low-density-of-states material like Mo_3Ge will rise with disorder while the high $n(E_F)$ in Nb_3Sn will drop, in a similar manner to the pure metals.

We now turn to the discussion of electronic transport in Mo_3Ge. At low temperatures, in contrast to the high-T_c $A15$ superconductors where a T^2 behavior is observed, a T^5 behavior is found and Matthiesson's rule is followed for initial damage levels. In Fig. 2 we show this data and also the "saturation" which occurs in a substance where λ is presumably increasing. A computer analysis of the data also shows that the T^5 behavior is preserved with initial damage, although with a decreasing coefficient. Then as the samples are successively disordered the behavior appears to change to a lower power of T.

For high temperatures the usual theory of electronic transport gives

$$\rho = \rho_0 + \frac{2\pi k_B T}{\hbar (n/m)_{\text{eff}} e^2} \lambda_{\text{tr}},$$

where the effective ratio $(n/m)_{\text{eff}}$ is given by the

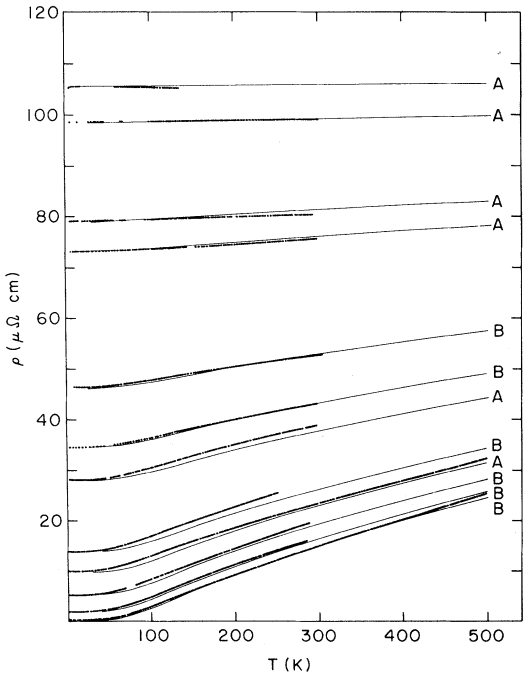

FIG. 2. ρ vs T as two Mo_3Ge samples are consecutively damaged. A, sputtered film; B, electron-beam-evaporated film. Solid lines are fitted by Eq. (1) using $\rho_{\max} = 120$ $\mu\Omega$ cm and $\rho_{\text{ideal}} = \rho_{0i} + LTG(\Theta_D/T)$, where $G(\Theta_D/T)$ is the Grüneisen function and Θ_D is taken to be 435 K. $L = C\lambda$ is determined by fitting the data for sample B when $\rho_0 \sim 0.15$ $\mu\Omega$ cm.

averaged Fermi velocity squared so that

$$\sum_k V_{k,x}{}^2 \delta(E_k - E_F) = N(E) \langle V_x{}^2(E_F) \rangle$$

and λ_{tr} is the transport electron-phonon coupling constant. In a qualitative discussion it is legitimate to replace λ_{tr} by λ the electron-phonon coupling parameter which determines T_c. From the increase of T_c with increasing ρ_0 we conclude that λ is also increasing. Since $n(E)$ is high when the average velocity on the constant energy surface $E = E_k$ is small and small when $V_x(E_F)$ is large we do not expect $(n/m)_{eff}$ to change much with increasing $n(E)$.[5] Thus on the basis of this conventional picture we would expect that the coefficient of the linear term at high temperatures should rise with ρ_0. Instead, as is shown in Fig. 2, the slope of ρ vs T curve decreases by a factor of 10 as ρ_0 changes from 0.05 to ~90 $\mu\Omega$ cm, indicating that a new saturation mechanism is at work. In fact, if we assume that for mean free paths of the order of an interatomic spacing an unspecified new channel of conducting becomes important[9,10] and write

$$\rho = (1/\rho_{ideal} + 1/\rho_{max})^{-1}, \quad (1)$$

where $\rho_{ideal} = \rho_0 + LTG(\Theta_D/T)$ is the resistivity as calculated in the conventional theory, $\rho_{max}{}^{-1}$ represents the conductivity of the new channel, and $G(\Theta_D/T)$ is the Grüneisen function,[11] then the theoretical curves for ρ in Fig. 2 can be explained with the single values of $\rho_{max} = 120$ $\rho\Omega$ cm and $\Theta_D = 435$ K. $L = C\lambda$ is determined by fitting the data for the best sample and for subsequent irradiations λ is determined from T_c via McMillan's equation. T_c is assumed to be an empirical function of ρ_0, i.e., $T_c = 1.45 + 0.3\rho_0$. Since λ only changes about 25% over the range of T_c changes, this is a relatively insensitive correction.

The reasonable fit to the data using a single value of ρ_{max} and the above procedure for determining λ suggests that the usual Boltzmann equation leaves out something very specific from consideration. Recently Chakraborty and Allen[10] have generalized the Boltzmann theory to include an interband conduction channel, and this theory shows some promise for understanding the parallel resistor form of Eq. (1). Of course, we have no particular reason to believe that Eq. (1) is the only valid formula for saturation, and recently Cote and Meisel[12] have suggested a somewhat different result based on the idea of phonon ineffectiveness.

We speculate that the physical source of the saturation has to do with the fact that electrons with short mean free path, l, "see" their environment as an inhomogeneous conductor (on the scale of l) and avoid the highly resistive regions. To make this idea into a phenomenological theory we note that the effective resistivity of a weakly inhomogeneous resistor is $\rho_{eff} = \bar{\rho}[\frac{2}{3}\langle(\Delta\rho)^2\rangle/\bar{\rho}^2]$,[13] where $\langle(\Delta\rho)^2\rangle$ is the mean-square fluctuation of ρ about its average $\bar{\rho}$. If we now assume that the width of the distribution of ρ is inversely proportional to the volume within which ρ is not fluctuating, e.g., l^3 and write that $\langle(\Delta\rho)^2\rangle \propto l^{-3} \cong \beta\rho^{-3}$, from $\rho_{eff}{}^{-1} = \bar{\rho}^{-1} + \frac{2}{3}\bar{\rho}^{-1}[\langle(\Delta\rho)^2\rangle/\bar{\rho}^2]$ we get Eq. (1) with $\bar{\rho} = \rho_{ideal}$ and $\rho_{max}{}^{-1} = \frac{2}{3}\beta$, a material parameter. Note that we have obtained ρ_{eff} in the form $\rho_{eff} = al^{-1} + bl^{-2}$. Evidently bl^{-2} is the first correction to the Boltzmann-equation result.

In summary, we present the following picture for the $A15$ compounds.

(a) In radiation-damaged $A15$ compounds, the two-peaked curve[14] of T_c vs e/a (electron-per-atom ratio) turns into a single-peaked empirical correlation centered at $e/a \approx 6$ just as the two-peaked curves of T_c vs e/a for the bcc transition metals becomes a single-peaked curve for the amorphous metals containing the same elements.[15]

(b) This behavior may be understood by assuming, as is conventional, that high $n(E_F)$ implies high T_c and that low $n(E_F)$ means low T_c, and by supposing that large amounts of disorder "smear" out the band structure in the sense that high $n(E_F)$ falls and low $n(E_F)$ rises with increasing disorder. The changes in T_c in the $A15$'s are expected to follow these changes in $n(E_F)$ in a manner similar to that proposed for the bcc transition metals.[15,16]

(c) Unlike the changes in the superconducting properties, the variation of the resistivity with increasing level of damage cannot be understood in conventional microscopic terms involving the Boltzmann equation. Phenomenologically, it can be viewed as further evidence for the existence of a saturation mechanism[9,17] which does not appear to reduce the electron-phonon interaction.

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Direct Observation of the Electronic Mean Free Path in Metallic Systems

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Conduction electrons injected from a ferromagnet F into a normal metal M can be experimentally distinguished from the intrinsic conduction electrons of M. They contribute to the anomalous Hall conductance in M, which allows a direct observation of their propagation in M. I develop a theory of this effect and apply this new method to quench-condensed In.

The propagation of conduction electrons in a metallic system determines its electronic transport properties. In this paper I present a new method to observe this propagation. The experimental procedure is in principle the following. On top of a metallic ferromagnet F, I condense the metal of interest, M. The conduction electrons enter from F into M. These electrons can be distinguished from the intrinsic electrons of M and it is possible to observe their penetration into M subsequently.

For a heuristic understanding one can choose the coordinate system with the z direction perpendicular to the interface between F and M. The ferromagnetic metal is magnetized in the z direction. In addition, an electrical field is applied parallel to the interface in the x direction. It is characteristic for a ferromagnetic metal that the current does not flow parallel to the electric field in the x direction but possesses an additional y component.[1] $I_y/I_x = \tan\varphi$ determines the Hall angle. As a consequence the electrical field does not only shift the entire Fermi surface in the k_x direction but also in the k_y direction. Electrons of the upper part of this asymmetrically shifted Fermi surface ($v_z > 0$) enter into the metal M. Normally in M the current flows only parallel to the applied electrical field $\vec{E}$ in the x direction. However, the conduction electrons which entered from F contribute also to a current in the y direction. Therefore, by measuring the off-diagonal conductance L_{xy} as a function of the thickness of M it is possible to obtain directly a picture of the path and penetration of the conduction

