

shock heated to  $10^{-2}$  keV, then rides up the  $\gamma = \frac{5}{3}$  adiabat to 2 keV, where the final shock collapse and burn raises  $T_i$  to 95 keV, prior to the expansive disassembly of the core. Clearly, the expansion pursues a similar adiabat down to  $\rho = 0.1$  g/cm<sup>3</sup>.

Optimal yield ratios (from pulses using  $\dot{E}_0^*$ ) are given versus  $\dot{E}_{in}^*$  in Fig. 2(e). Here the calculations for spheres employed local  $\alpha$  deposition, which artificially improves their performance at small  $m$ . For the shells with nonlocal, time-dependent deposition, the rules  $Y_R^* \sim (E_{in}^*)^{0.45}$ ,  $E_{in}^* > 8$  kJ, and  $Y_R^* \sim (E_{in}^*)^{0.90}$ ,  $E_{in}^* \leq 8$  kJ, are observed. We calculate  $Y_R^* = 4.7$  for only 1.9 kJ delivered to a 3- $\mu$ g shell. Thus, breakeven with only 350 J of input energy is implied. With non-local deposition and parameters other than  $R$  and  $\dot{E}_0^*$  held constant, our studies show degraded performance for shells relative to spheres under the exposure profile (1), as documented by Fig. 2(f).

Thus, within the constraints of the physical model described, our calculations imply the feasibility of controlled thermonuclear burn by inertial confinement. Scaling laws for a broad range of potential target designs have been presented. Still numerous questions remain, as to the three-dimensional stability of these designs, the details of anomalous light absorption,<sup>13</sup> hyperthermal electron production and deposition,<sup>14</sup> and the effects of spontaneously generated magnetic fields.<sup>15</sup>

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<sup>1</sup>K. Boyer, Bull. Amer. Phys. Soc. **17**, 1019 (1972).

<sup>2</sup>S. Glasstone and R. H. Lovberg, *Controlled Thermonuclear Reactions* (Van Nostrand, Princeton, N.J., 1960), p. 33.

<sup>3</sup>J. L. Tuck, Nucl. Fusion **1**, 202 (1961).

<sup>4</sup>M. S. Chu, Phys. Fluids **15**, 413 (1972).

<sup>5</sup>As described in D. Hawkins, LASL Report No. LAMS-2532, 1961 (unpublished), Vol. I, p. 23.

<sup>6</sup>J. Nuckolls, L. Wood, A. Thiessen, and G. Zimmerman, Nature (London) **239**, 139 (1972).

<sup>7</sup>J. P. Silin, Zh. Eksp. Teor. Fiz. **48**, 1679 (1965) [Sov. Phys. JETP **21**, 1127 (1965)].

<sup>8</sup>P. K. Kaw and J. M. Dawson, Phys. Fluids **12**, 2586 (1969).

<sup>9</sup>J. P. Freidberg and B. M. Marder, Phys. Rev. A **4**, 1549 (1971).

<sup>10</sup>J. P. Freidberg, R. W. Mitchell, R. L. Morse, and L. I. Rudinski, Phys. Rev. Lett. **28**, 795 (1972).

<sup>11</sup>J. L. Bobin, Phys. Fluids **14**, 2341 (1971).

<sup>12</sup>R. S. Cooper, LASL Report No. LA-DC-72-104, 1972 (unpublished).

<sup>13</sup>D. W. Forslund, J. M. Kindel, and E. L. Lindman, Bull. Amer. Phys. Soc. **17**, 1044 (1972).

<sup>14</sup>R. L. Morse and C. W. Nielson, "Occurrence of High-Energy Electrons and Surface Expansion in Radically Heated Target Plasmas" (to be published).

<sup>15</sup>J. A. Stamper, K. Papadopoulos, R. N. Sudan, S. O. Dean, E. A. McLean, and J. M. Dawson, Phys. Rev. Lett. **26**, 17 (1971).

## Superconductivity in "Amorphous" Transition-Metal Alloy Films

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The superconducting transition temperatures of "amorphous" transition-metal alloys have been measured. The variation of  $T_c$  as a function of the electron-to-atom ratio is consistent with the presence of an atomlike parameter characterizing the systematics of  $T_c$ , with, however, an unexplained peak at the half-filled  $d$  shell. The data do not support a recent explanation based entirely on a simple averaging or smearing of the crystalline electronic density of states.

In this Letter we report measurements of the superconducting transition temperature  $T_c$  for disordered  $4d$  and  $5d$  transition-metal alloy films.<sup>1</sup> We have observed that the  $T_c$  in amorphous (or highly disordered) transition metals

differs strongly from that of crystalline transition metals (see Figs. 1 and 2). The resulting behavior of the amorphous  $T_c$  is believed to be a manifestation of "the dominance of atoms and their local environment in determining supercon-

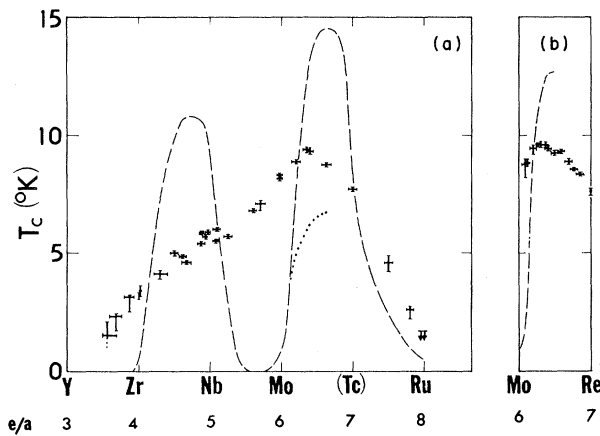


FIG. 1. Superconducting transition temperatures of vapor-quenched (a)  $4d$  transition-metal alloy and (b) a  $4d$ - $5d$  alloy are shown as a function of the number of electrons per atom ( $e/a$ ). These data are consistent with the presence of an atomlike parameter controlling  $T_c$  in the amorphous or disordered transition metals. The corresponding crystalline  $T_c$  data are indicated by the dashed curve (dotted curve for the Mo-Ru alloys). In (a) the amorphous data are for Y-Zr, Nb-Zr, Mo-Zr, Nb-Mo, and Mo-Ru alloys. At  $e/a=6$  to 7 the crystalline data (dashed curve) represents Mo-Tc and Mo-Re alloys. The amorphous data in (a) for  $e/a=6$  to 7 are to be compared with the lower dotted curve. The Mo-Re crystalline data are again reproduced in (b) for comparison with the amorphous Mo-Re films. The arrows indicate that the  $T_c$  are less than  $1.7^\circ\text{K}$ . The transition widths indicated were determined by the 10 to 90% of normal resistance interval; the compositions ( $e/a$ ) were determined by microprobe analysis.

ducting parameters . . .,"<sup>2</sup> and indicates the existence of an atomlike parameter that characterizes the systematics of superconductivity in the transition-metal series. An atomic parameter of this nature was first introduced by Hopfield<sup>2</sup> and has recently been discussed further by Benemann and Garland,<sup>3,4</sup> who predict a smooth maximum in the region 7 to 8 electrons per atom. The data presented here, when interpreted in terms of this parameter  $\eta$ , indicate that instead  $\eta$  has an unexplained sharp triangular peak in the middle of the  $d$  series. The present explanation of the data, in terms of the Hopfield or similar atomlike parameter, differs from a recent explanation of the  $T_c$  in disordered transition metals based entirely upon a simple smearing or averaging of the electronic density of states.<sup>5,6</sup>

Past studies of amorphous transition metals,<sup>5-8</sup> motivated by the possibility of higher  $T_c$ 's via lattice disorder, have not been as extensive as that presented here. This study was stimulated

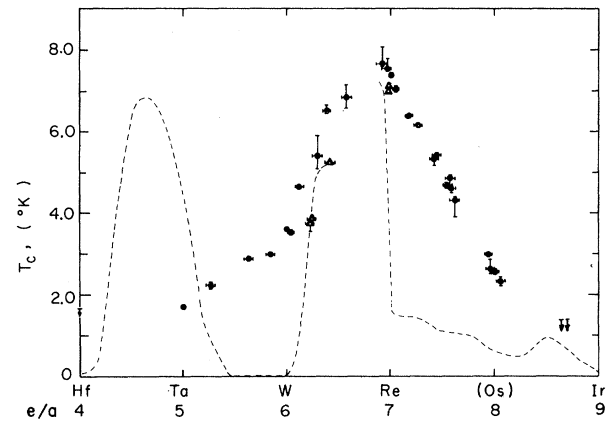


FIG. 2. Superconducting transition temperatures of vapor-quenched  $5d$  transition-metal alloys are shown as a function of  $e/a$ . Corresponding crystalline  $T_c$  data are indicated by the dashed curve. Amorphous  $T_c$  data are for the  $5d$  elements and Ta-W, Ta-Re ( $\Delta$ ), W-Re, and Re-Ir alloys.

by our previous data,<sup>8</sup> which suggested that there may be a regular variation of  $T_c$  across the amorphous transition-metal series. A similar suggestion by Gamble and Geballe appeared at about the same time.<sup>7</sup>

The superconducting transition temperatures of the amorphous films of the  $4d$  and  $5d$  transition-metal alloys are shown in Fig. 1(a) and Fig. 2. The primary feature of the data is that  $T_c$  increases as a function of the number of electrons per atom ( $e/a$ ) in a smooth, nearly linear fashion to a definite triangular peak near the middle of the  $d$  series. The peak occurs for the  $4d$  series at  $e/a=6.4$  in contrast to the crystalline maximum at 6.7.  $T_c$  then decreases at nearly the same slope, followed by a more rapid decrease at  $e/a \sim 7$ . In the  $5d$  series, the amorphous  $T_c$  peak occurs near  $e/a=6.8$ ; the crystalline maximum is not well established in the  $5d$  series. The smooth behavior in  $T_c$  across two phase regions of the alloy systems studied demonstrates that  $e/a$  determines  $T_c$  in the "amorphous" state, independent of the normal crystal structure. Furthermore, the amorphous  $T_c$  curve for Mo-Re, shown in Fig. 1(b), is nearly identical to the amorphous Mo-Ru at the same  $e/a$  ratio, while the corresponding crystalline  $T_c$  for these alloys are markedly different.

The metal films were prepared by electron beam evaporation onto a sapphire substrate at  $4.2^\circ\text{K}$  in a vacuum of  $8 \times 10^{-8}$  to  $1 \times 10^{-7}$  Torr, and at rates of  $50$ - $300 \text{ \AA}/\text{sec}$ . Gas impurities ( $\text{O}_2$ )

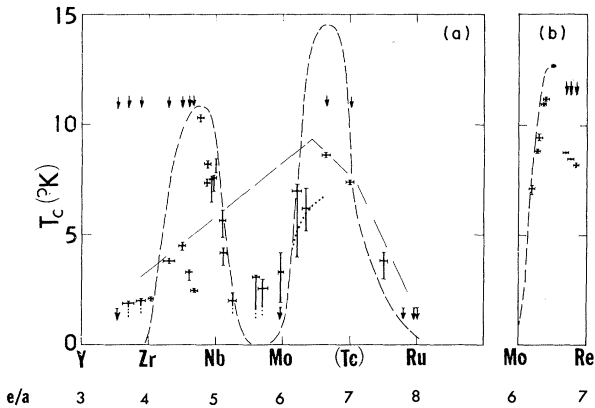


FIG. 3. Superconducting transition temperatures of (a) 4d transition-metal alloys and (b) Mo-Re alloys after warming to 300°K. The  $T_c$ 's generally returned to or toward their corresponding crystalline values (dashed and dotted curves) and had exhibited large irreversible decreases in resistivity during the warming cycle. The exceptions, indicated by arrows along the upper portion of the figure, were found to be "amorphouslike" when examined by electron diffraction (see text).

up to  $3 \times 10^{-6}$  Torr produced no significant change in the amorphous  $T_c$ .  $T_c$  was measured resistively<sup>9</sup> *in situ* and the films were then warmed to 300°K, cooled back down to 4.2°K, and  $T_c$  was re-measured. Film composition was determined by electron microprobe analysis resulting in an accuracy of  $\pm 3$  at.% for 4d alloys,  $\pm 5$  at.% for Mo-Re, and  $\pm 4$  at.% for the 5d alloys. Experimental aspects will be discussed in detail later.<sup>10</sup>

Figure 3 shows the values of  $T_c$  for the 4d series and Mo-Re measured following the warming to 300°K and then recooling. Similar results were found for the 5d series (not shown). Except for a few cases, the  $T_c$ 's have changed toward the bulk crystalline value. Those exceptions, indicated by the arrows, also did not exhibit significant change in resistivity during the temperature cycling from 4 to 300°K and back. In fact, the annealing behavior, subsequent  $T_c$ , and electron diffraction data form a coherent picture as follows: Those alloys for which there was an annealing step (a relatively sharp change in resistivity indicative of a change in phase of the bulk of the sample) also showed a change in  $T_c$  towards the bulk crystalline  $T_c$ , and the electron diffraction and electron micrographs indicated the normal crystal structure with grain sizes ranging from 50 to 1000 Å. On the other hand, for those films for which the  $T_c$  and resistivity did not change, electron diffraction pictures re-

vealed diffuse three- or four-ring patterns, characteristic of liquid or amorphous materials. In addition, electron micrographs revealed no grain structure or other inhomogeneity to the limit of resolution of the microscope ( $\approx 20$ – $25$  Å). On the basis of the above observations we infer that all our specimens were prepared in an "amorphous" state<sup>11</sup> and those with compositions corresponding to equilibrium two-phase regions remain in this state up to room temperature and above. The annealing and electron microscopy results are discussed more fully elsewhere.<sup>10,12</sup>

That the measurements reported here are consistent with the behavior of the Hopfield parameter  $\eta$  or a similar atomiclike parameter<sup>13</sup> is seen by considering the electron-phonon interaction parameter  $\lambda$  as discussed by McMillan<sup>14</sup>:  $\lambda = N(0)\langle I^2 \rangle / M\langle \omega^2 \rangle$ , where  $M$  is the atomic mass,  $\langle \omega^2 \rangle$  is the mean squared phonon frequency,  $N(0)$  is the electronic density of states, and  $\langle I^2 \rangle$  is the squared averaged electron-phonon matrix element. McMillan noted that  $N(0)\langle I^2 \rangle$  is approximately constant for the bcc transition metals, which Hopfield<sup>2</sup> explained by deducing that it is a "local atomic parameter,"  $\eta \approx N(0)\langle I^2 \rangle$ , independent of  $N(0)$  and varying smoothly across the transition-metal series. Recent calculations have modified the meaning of  $\eta$  from a strictly atomic parameter to an overlap integral among the atoms,<sup>3,4</sup> and, as discussed by Barisić *et al.*,<sup>15</sup> to the  $d$ -electron contribution to the cohesive energy  $E_c^d$ . The large variation in  $1/\langle \omega^2 \rangle$  dominates the behavior of  $\lambda$  and  $T_c$  in the crystalline state.<sup>14</sup> Since the crystalline  $N(0)$  exhibits a still unexplained strong correlation with  $1/\Theta_D^2$  (and therefore  $1/\langle \omega^2 \rangle$ ), it is believed to be the factor controlling  $\lambda$  and  $T_c$  in the crystalline state.<sup>2,14,16</sup> In the amorphous state the  $N(0)$  is assumed to be smoothed out versus  $e/a$  by the disorder,<sup>4-6</sup> allowing the variation with  $e/a$  of the amorphous  $T_c$  to be determined by the parameter  $\eta$  and/or other atomiclike parameters.

We believe our data are the first experimental evidence that shows features characteristic of an atomiclike local parameter and its variation across the transition-metal series.<sup>17</sup> An unexpected feature is the indication of a triangular peak in the middle of the  $d$  band instead of a smooth peak at a larger  $e/a$ .<sup>3</sup> On the basis of the calculation of Barisić, Labbé, and Friedel<sup>15</sup> relating  $\eta$  to  $E_c^d$ , and of an examination of the bulk cohesive energy curve,<sup>18</sup> we judge that the peak occurs at the half-filled  $d$  shell, i.e., an integral number (five) of  $d$  electrons per atom.

This, however, is not always at an integral number of the total  $e/a$ .

The amorphous  $T_c$  data cannot be explained solely on the basis of a smearing of the density of states by the disorder, as has been proposed.<sup>5,6</sup> This argument states that because of a short mean free path in the disordered alloys the electron energy states are broadened, and the sharp structure in the density of states characteristic of the transition-metal series is averaged over energy. This results in those alloys with a high  $T_c$  in the crystalline state having a lower  $T_c$  in the disordered state, and vice versa for those alloys with a low  $T_c$ . However, this argument fails to explain (i) the higher  $T_c$  for the amorphous Mo-Ru and W-Re alloys compared to their crystalline counterparts near the maximum in the crystalline  $N(0)$ , and (ii) the sharp triangular peak in the amorphous  $T_c$  and the position of the peak.

In conclusion, the  $T_c$  data presented here are consistent with the presence of an atomiclike parameter characterizing the systematics of  $T_c$  in transition metals. A feature of the data that is not explained by recent theories is the sharp peak at a half-filled  $d$  shell.

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<sup>1</sup>Preliminary results were given in M. Collver *et al.*, Bull. Amer. Phys. Soc. **15**, 1613 (1970). Also see R. H. Hammond and M. M. Collver, Lawrence Radiation Laboratory, University of California, Berkeley, Report No. UCRL-20500, 1970 (unpublished), p. 137; and M. M. Collver, Ph. D. thesis, Lawrence Berkeley Laboratory Report No. LBL-167, 1971 (unpublished).

<sup>2</sup>J. J. Hopfield, Phys. Rev. **186**, 443 (1969).

<sup>3</sup>K. H. Benneman and J. W. Garland, in *Superconductivity in d- and f-Band Metals*, AIP Conference Pro-

ceedings No. 4, edited by D. H. Douglass (American Institute of Physics, New York, 1972), p. 103.

<sup>4</sup>J. W. Garland and K. H. Benneman, in *Superconductivity in d- and f-Band Metals*, AIP Conference Proceedings No. 4, edited by D. H. Douglass (American Institute of Physics, New York, 1972), p. 255.

<sup>5</sup>J. E. Crow, M. Strongin, R. S. Thompson, and O. F. Kammerer, Phys. Lett. **30A**, 161 (1969).

<sup>6</sup>M. Strongin, in *Superconductivity in d- and f-Band Metals*, AIP Conference Proceedings No. 4, edited by D. H. Douglass (American Institute of Physics, New York, 1972), p. 223.

<sup>7</sup>F. R. Gamble and T. H. Geballe, Bull. Amer. Phys. Soc. **15**, 343 (1970).

<sup>8</sup>M. M. Collver, J. A. Roberts, and R. H. Hammond, UCRL Report No. UCRL-19155, 1969 (unpublished), p. 134.

<sup>9</sup>We do not believe errors have arisen because of, say, superconducting filaments in the bulk of the film or along their edges. The evidence against such problems include (a) the homogeneity of the samples as shown by electron-microscope examination, and sharpness of the amorphous-to-crystalline phase transition; (b) edges removed from unannealed samples did not change  $T_c$ .

<sup>10</sup>M. M. Collver and R. H. Hammond, to be published.

<sup>11</sup>We note that our interpretation of the results does not depend on the exact meaning of the word "amorphous": Very short mean free path due to disorder is sufficient.

<sup>12</sup>R. Loop, M. Collver, and R. Hammond, in *Electron Microscopy and Structure of Materials*, edited by G. Thomas *et al.* (Univ. of California Press, Berkeley, 1972); R. Loop, M. S. thesis, Lawrence Berkeley Laboratory Report No. LBL-177, 1971 (unpublished).

<sup>13</sup>Although the analysis of the data in terms of  $\eta$  is the most reasonable explanation, and probably explains the general features observed, it cannot be ruled out that other atomiclike factors could play a role and, for example, give rise to the sharpness in the observed peak. These include the screened Coulomb interaction  $V_c$  [see W. L. McMillan, Phys. Rev. **167**, 331 (1968)], where a dip at the half-filled  $d$  shell would explain the peak in  $T_c$ . There is at the present time no reason to expect that this will occur.

<sup>14</sup>McMillan, Ref. 13.

<sup>15</sup>S. Barisic, J. Labbé, and J. Friedel, Phys. Rev. Lett. **25**, 919 (1970).

<sup>16</sup>F. Heiniger, E. Bucher, and J. Muller, Phys. Kondens. Mater. **5**, 243 (1966).

<sup>17</sup>See Ref. 4 for an analysis of a similar behavior found in  $U_6X$  alloys.

<sup>18</sup>R. E. Watson and H. Ehrenreich, Commun. Solid State Phys. **B 3**, 109 (1970); K. A. Gschneider, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1964), Vol. 16, p. 275.