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SUPERCONDUCTIVITY OF A La-Zn 22 AT % AMORPHOUS ALLOY

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ABSTRACT - We report here the main results of superconductivity measurements on a La-Zn 22 at % alloy, in amorphous and crystalline (hexagonal metastable phase) forms. The experimental details and a complete discussion of these results will be published elsewhere. The superconducting and relevant electronic properties have been extracted from specific heat, upper critical field and electrical resistivity measurements. The difference in the superconducting behaviour between the amorphous and crystalline states is compared to theoretical predictions. In conjunction with other results those presented here throw light on the problem of the electron-phonon coupling strength in amorphous transition metals alloys.

I. PREPARATION OF THE SAMPLES. The amorphous samples are obtained in the form of thick films (thickness about 100 µm) by high rate sputtering of a bulk pre-alloyed target at the eutectic composition (23 at % Zn). Condensation occurs on a substrate held at liquid nitrogen temperature. The sample weight is about 4 g and the chemical composition is 21.8 at % Zn. The X-ray diffraction spectrum is shown in fig. 1a. The single broad peak is located at \( \theta = 30.9° \). The nearest-neighbour distance (nnd) calculated by the Debye formula \( \theta = \frac{\pi a}{\theta_m} \) is 3.56 Å. The crystallisation of the amorphous alloy has been studied as a function of temperature and time. For the superconductivity studies, the sample was subjected to a thermal treatment of 2 minutes at 90°C followed by 2 minutes at 180°C. This treatment lead to a single hexagonal metastable phase with \( a = 6.48 \) Å and \( c/a = 0.593 \). The corresponding X-ray diffraction spectrum is shown in fig. 1b. A consideration of the number of atoms in a unit cell of this phase and the atomic volumes of La and Zn, indicates that there is no long-range order for the Zn atoms. The chemical disorder therefore must be of the same type in the amorphous and crystallised samples. Subsequently we shall designate this crystalline sample by hex-LaZn.

II. RESULTS OF THE LOW TEMPERATURE MEASUREMENTS. The specific heat has been measured from 0.06 K to 6.5 K for a-La-Zn and hex-La-Zn. Experimental details, the \( C_p(T) \) curves and a discussion of the very low temperature part of the \( C_p(T) \) curves are given in a separate paper /2/. The superconducting parameters and relevant quantities extracted from the \( C_p(T) \) data are summarized in table I. The symbols used here have the conventional meaning and the values have been obtained by the usual methods /3/. The superconducting electronic specific heat \( C_{es} \), obtained
by subtracting the "lattice" $T^3$ term from the total specific heat $C_p(T)$ below $T_c$, is plotted in fig. 2.

The electrical resistivity has been measured between $T_c$ and 300 K by a 4-probes ac technique. The upper critical field $H_{c2}$ measurements have been carried out by the same method as the resistivity with a transverse magnetic field supplied by a superconducting magnet. For a-LaZn and hex-LaZn the measurements have been performed in liquid helium between 1.7 K and $T_c$. For a-LaZn, a second sample (with a slightly different $T_c$) has been measured down to 0.135 K in an adiabatic demagnetization apparatus.

The experimental results are shown in fig. 3 and the reduced thermal variations of $H_{c2}$ for the two a-LaZn samples are plotted on fig. 4. Table II summarizes the low temperature electronic transport and electromagnetic properties. In this table we have also reported the theoretical value of $H'_{c2}(T_c)$ calculated from the BCS-Gorkov theory $\Delta_0^f$.

### Table I

<table>
<thead>
<tr>
<th>Alloy</th>
<th>$T_c$ K</th>
<th>$\Theta_d$ K</th>
<th>$\gamma$</th>
<th>$\Delta_c(T_c)$ kOe K$^{-1}$</th>
<th>$H_{c2}(0)$ kOe</th>
<th>$\Delta c'/\Theta_d$, ($T=T_c$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a-LaZn$_{0.22}$</td>
<td>3.15</td>
<td>96</td>
<td>7.9</td>
<td>0.36</td>
<td>0.63</td>
<td>2.60</td>
</tr>
<tr>
<td>hex-LaZn$_{0.22}$</td>
<td>2.23</td>
<td>110</td>
<td>10.2</td>
<td>0.30</td>
<td>0.38</td>
<td>1.40</td>
</tr>
</tbody>
</table>

$k_B^2(T_c) = |dH_{c2}/dT|_{T=T_c}$. 

### Table II

<table>
<thead>
<tr>
<th>Alloy</th>
<th>$\rho(T_c)$ $\mu\Omega$-cm</th>
<th>$H_{c2}(T_c)$ kOe K$^{-1}$</th>
<th>$H_{c2}(0)$ kOe</th>
<th>$H'_{c2}(T_c)$ BCS kOe K$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>a-LaZn$_{0.22}$</td>
<td>150</td>
<td>26.0</td>
<td>71.0</td>
<td>26.1</td>
</tr>
<tr>
<td>hex-LaZn$_{0.22}$</td>
<td>135</td>
<td>16.5</td>
<td>-</td>
<td>29.9</td>
</tr>
</tbody>
</table>

$k_B^2(T_c) = |dH_{c2}/dT|_{T=T_c}$. 

fig. 3

fig. 4

LaZn 22% at

- sample 1
- sample 2
- amorphous
- crystalline sample(hex)

$T_c(1)=4.12 K$
$T_c(2)=4.18 K$

$H_{c2}(T)/H_{c2}(0)$

$\Delta_0/kT_c = 2.1$

$\Delta_0/kT_c = 1.45$

$\rho(T_c)$ $\mu\Omega$-cm

$H_{c2}(T_c)$ kOe K$^{-1}$

$H_{c2}(0)$ kOe

$H'_{c2}(T_c)$ BCS kOe K$^{-1}$
III. DISCUSSION. Electron-phonon coupling and $T_c$

From the values of $\Delta C/\gamma T_c$ and $\Delta p/k_B T_c$ (table I) compared to the BCS values, it appears that a-LaZn is a strong coupling superconductor and hex-LaZn, a weak one. If we assume (see §1) that the chemical order is about the same for the amorphous and crystalline phases, the main difference between the two phases lies in the topological order. We can therefore conclude that topological disorder alone increases the electron-phonon coupling. This experimental fact is in agreement with the theoretical predictions of Bergmann /5/ that the lack of translational invariance in amorphous metals provides an additional phase space for electron-phonon coupling. We can see also from the table I that $\theta_D$ and $N(0)$ (or $\gamma$) increase from the amorphous to the crystalline phases; we can therefore associate the observed decrease of $T_c$ with the decrease of the coupling. Such a decrease of $T_c$ during the early stages of crystallisation, has already been observed in Zr-Rh /6/ and Zr-Pd /7/ alloys.

Upper critical field $H_{c2}$. It can be seen from table II that the experimental value of $H_{c2}^* (T_c)$ for a-LaZn, agrees remarkably well with the theoretical value $H_{c2}^* (T_c)_{BCS}$ calculated from the BCS-Gorkov theory of weak-coupling superconductors. Such a result has already been obtained /1/ by Rainer and Bergmann /8/ for amorphous $s$-$p$ metals who explained it by the characteristic shape of the Eliashberg function $\alpha^2F(\omega)$ at low frequencies, peculiar to strong-coupling amorphous metals /2/ by Shull et al /9/ for amorphous alloys LaGa$_{0.20}$ and LaGa$_{0.22}$, which are intermediate coupled superconductors with d-electrons. On the contrary, $H_{c2}^* (T_c)$ for hex-LaZn is different from the BCS value in spite of its weak coupling character. It is possible, for this badly crystallized sample with very small crystallites, that the measured $\rho(T_c)$ does not represent the "intra-crystal" resistivity which can be much smaller. Finally, fig. 4 shows, for a-LaZn, that $H_{c2}^* (T)$ follows the theoretical thermal variation predicted by Maki /4/ with $\alpha = 0$ and $\lambda_{so} = \infty$. This agreement is perhaps fortuitous because in this alloy $\alpha$ is different from zero and $\lambda_{so}$ is unknown. This thermal behaviour is different from that found for others amorphous alloys /8, 10, 11, 12/ where $H_{c2}^*(T)$ decreases faster than the Maki law.

Comparison with other amorphous alloys of transition metals. In table III we show some published results on amorphous alloys where $C_p(T)$, $\rho_o = \rho(T_c)$ and $H_{c2}^* (T)$ have been simultaneously measured. It can be seen from this table that the electron-phonon coupling (expressed by $\Delta C/\gamma T_c$) increases from one alloy to another when $\rho(T_c)$ decreases. This behaviour is in qualitative agreement with a theory recently established by Meisel and Cote /13/ to calculate the decrease of $T_c$ with increasing $\rho_o$, as observed in highly resistive crystalline alloys (Nb and Al). When the electronic mean free path, $\xi$, is small enough, the electron-phonon coupling decreases with $\xi$. This results from the Zimmann-Pippard condition for the electron-phonon diffusion ($2\pi q_{ph} < \xi$) which introduces a $\xi^{-1}$ low-frequency cut-off in the Eliashberg function $\alpha^2F(\omega)$ and causes a decrease of the coupling strength. If we assume that the variations of $\rho(T_c)$ reflect the variations of $\xi$ for the alloys of table III ($N(0)$ varies only a little) and that $\alpha^2F(\omega)$ is about the same, then the

<table>
<thead>
<tr>
<th>Alloy</th>
<th>$T_c$</th>
<th>$\theta_D$</th>
<th>$N(0)$</th>
<th>$\Delta C/\gamma T_c$</th>
<th>$\rho(T_c)$</th>
<th>$H_{c2}^* (T_c)$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>a-LaZn, 0.22</td>
<td>4.15</td>
<td>96</td>
<td>3.1</td>
<td>10$^{-9}$</td>
<td>2.60</td>
<td>150</td>
<td>-</td>
</tr>
<tr>
<td>a-LaZn, 0.22</td>
<td>3.64</td>
<td>109</td>
<td>3.2</td>
<td>10$^{-9}$</td>
<td>1.96</td>
<td>200</td>
<td>22.5</td>
</tr>
<tr>
<td>a-LaZn, 0.24</td>
<td>3.28</td>
<td>99</td>
<td>3.1</td>
<td>10$^{-9}$</td>
<td>1.7</td>
<td>227</td>
<td>24.0</td>
</tr>
<tr>
<td>a-LaZn, 0.22</td>
<td>2.53</td>
<td>180</td>
<td>3.0</td>
<td>10$^{-9}$</td>
<td>1.61</td>
<td>246</td>
<td>26.5</td>
</tr>
<tr>
<td>(9) Calculated with an estimated density.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
electron-phonon coupling will decrease with increasing $\rho(T_C)$. More generally this model could qualitatively explain why some other amorphous alloys of transition metals have been found to be weak-coupling superconductors /14/. Likewise, such an inverse relation between the coupling strength and $\rho(T_C)$, associated with small variations of $N(O)$, could explain the relative constancy of $H'_c(T)$ observed in table III and for some fifteen other amorphous transition alloys /12,15/.

IV. CONCLUSION. A-LaZn$_{0.22}$ is a strong-coupling superconductor. After crystallisation into a metastable hexagonal phase the alloy becomes a weak coupled superconductor with a reduction of $T_C$ by a factor of about 2 (amorphous $T_C = 4.15$ K, crystallised $T_C = 2.23$ K). The increase of coupling with topological disorder, theoretically predicted by Bergmann, is verified for this d-alloy. The experimental $\left[\frac{dH'_c}{dT}\right]_{T=T_C}$ value is in agreement with the value calculated by the BCS-Gorkov theory of weak coupled superconductors, such as found by Bergmann for strong coupled amorphous s-p superconductors. The present results on a-LaZn, together with those obtained with other amorphous La-based alloys and a-PdZr alloy, indicate an inverse relation between the residual resistivity and the coupling strength. Such a relation, theoretically introduced by Meisel and Cote using the Zimmann-Pippard condition for electron-phonon diffusion, could qualitatively explain the spread of results concerning the coupling strength in d-amorphous alloys and the relative constancy of $\left[\frac{dH'_c}{dT}\right]_{T=T_C}$ observed in numerous d-amorphous alloys. Other experimental results on different d-amorphous alloys and more precise theoretical calculations will be necessary to confirm this trend.

REFERENCES