THE INFLUENCE OF MAGNETIC ORDERING ON THE SUPERCONDUCTING PROPERTIES OF AMORPHOUS $(\text{Zr}_{1-x}\text{Er}_x)_3\text{Rh}$ ALLOYS

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ABSTRACT

The effect of Er impurities on the superconducting properties of an amorphous Zr_3Rh alloy has been studied. The Er impurities are found to exhibit magnetic behavior characteristic of a free Er^{3+} ion in a weak crystal field. Magnetic ordering of the Er moments is observed to occur at a temperature T_M which is proportional to the Er concentration. This ordering strongly influences superconductivity as evidenced by anomalous behavior in the concentration dependence of T_c . For concentrations near the critical Abrikosov-Gor'kov value superconductivity appears to be induced by magnetic ordering. The superconducting critical field $H_{c2}(T)$ is strongly effected by magnetic ordering and can be used to deduce information concerning the nature of the ordering. The results are analyzed in terms of theoretical models.

The discovery of magnetic ordering of rare earth (R.E.) ions in ternary superconducting $(\text{R.E.})_1\text{Rh}_4\text{B}_4$ compounds has generated much interest in the problem of superconductivity and magnetic ordering.^{1,2,3} These compounds are unique in that the magnetic state is characterized by long range ordering. In superconducting alloys containing dilute concentrations of rare earth ions in solid solution, magnetically ordered states also exist and have been studied in conjunction with superconductivity.^{4,5,6} Anomalous behavior of the concentration dependence of T_c near the critical Abrikosov-Gor'kov (AG) concentration was reported for several alloys of this type.^{6,7} Bennemann⁸, Entel and Klose⁹, and others^{10,11} have attempted to present a theoretical basis for understanding these anomalies.

Amorphous superconductors possess a highly disordered atomic arrangement which results in a small electron mean free path ℓ , a small superconducting coherence length ξ ¹², and a spatial attenuation of the range of the RKKY interaction.^{13,14} This communication deals with the magnetic and superconducting properties of amorphous alloys of the form $(\text{Zr}_{1-x}\text{Er}_x)_3\text{Rh}$ which have been obtained by liquid quenching¹⁵ over the range $0 \leq x \leq 1$. The binary alloy Zr_3Rh is a d-band superconductor with $T_c = 4.5$ K.¹⁶ In this matrix, Er enters in the Er^{3+} valence state and carries a magnetic moment. This state is shown to be weakly split by crystal field effects producing a magnetic ionic ground state which has at least the double degeneracy of a Kramer's ion.¹⁷ Magnetic measurements in the concentration range $0 \leq x \leq 0.25$ show that the Er ions undergo a magnetic ordering transition with an ordering temperature T_M proportional to the Er concentration as given by the formula $T_M = 9.8 X(\text{K})$. The linear dependence of T_M on X together with other

data suggest that this transition is of the general spin glass type. A coexistence of superconductivity and magnetic ordering over a broad range ($X < 0.16$) of Er concentrations is observed. Furthermore, superconductivity is noticeably enhanced in the region where $T_C \approx T_M$. For $0.11 < X < 0.14$, superconductivity appears to be induced by magnetic ordering. It is argued that this rather unique behavior is likely to be common for amorphous d-band superconductors containing magnetic rare earth ions in concentrations below the percolation threshold for ferromagnetism.

The superconducting properties of the alloys were studied by four point resistivity measurements over the temperature range from 0.3 K to 10 K as well as by ac inductance bridge measurements down to 1.2 K. The upper critical field H_{C2} was measured by taking the midpoint of the resistive transition as a function of external field H and T . Magnetization measurements were made with a Faraday magnetometer in fields ranging up to 75 kG. All samples were examined by x-ray diffraction scanning to ensure their amorphous nature. Samples showing any evidence of crystallization were not used in the measurements.

A phase diagram summarizing the data is shown in Fig. 1. The variation of the superconducting transition temperature as measured by the midpoint of resistive transition is shown. The figure also shows the width of the transitions which is typically 0.2 K as measured from the 25% to 75% points on the resistive transition. The transition is somewhat broader for alloys with high Er content. This broadening is thought to arise from statistical fluctuations in the local Er content of the matrix. When significant fluctuations occur on a spatial scale comparable to ξ , an intrinsic broadening of the transition results. The

Fig. 1. Phase diagram of magnetic and superconducting states in amorphous (Zr_{1-x}Er_x)₃Rh alloys. Solid lines are described in the figure. Dashed line is drawn for aid in visualizing the superconducting phase boundary in the composition range 0.10 < x < 0.16. Downward arrow with horizontal bar indicates an incomplete superconducting transition. Downward arrow with no bar indicates no superconductivity down to T = 0.3 K.

midpoint of the inductively measured transitions (down to 1.2 K) is also shown in the figure for comparison. For $x \geq 0.13$, the low field ac susceptibility shows a pronounced magnetic ordering transition which is characterized by a well defined susceptibility maximum. These data are shown in Fig. 2 for $x = 0.10, 0.15, 0.20$, and 0.225 . For $x = 0.10$, the magnetic transition is preempted by a superconducting transition at $T = 1.45$ K as seen in the figure so that the susceptibility maximum is not observed. The temperature, T_M , at which the maxima occurs was found to be proportional to the Er concentration of the matrix as also seen in Fig. 1. The susceptibility maxima and its linear dependence on Er content are characteristics of a spin glass type of magnetic ordering.^{18,19} Poon and Durand²⁰ have discussed magnetic ordering in similar amorphous alloys of the type $(\text{La}_{1-x}\text{Gd}_x)_{80}\text{Au}_{20}$ in terms of a magnetic-cluster spin glass model. The present case differs from that discussed by Poon and Durand in that crystal field effects are important. This can be seen by analyzing the dc magnetization arising from Er as a function of field and temperature, $M(H,T)$. A complete set of data for $(\text{Zr}_{.95}\text{Er}_{.05})_3\text{Rh}$ is shown in Fig. 3 for $3 \text{ K} < T < 100 \text{ K}$ and $0 < H < 75 \text{ kG}$. At high temperatures ($T > 10 \text{ K}$), the initial susceptibility is well described by a Curie-Weiss law $\chi = C/(T+\theta_p)$. For $x \approx 0.05$, the paramagnetic ordering temperature $\theta_p = 5.1 \text{ K}$. From the value of C obtained by a least squares fit, one can deduce the effective moment of Er in the paramagnetic regime. One obtains $\mu_{\text{Er}} = 9.88 \mu_B$ which is in good agreement with the value $9.59 \mu_B$ expected from Hund's rule for Er^{3+} in the $^4I_{15/2}$ electronic state. At low temperature, a significant deviation from the Curie-Weiss behavior is seen beginning at $T \approx 6 \text{ K}$. This is attributed to the effect of magnetic ordering as discussed later. The low temperature high-field

Fig. 2. Relative inductance as a function of temperature for several amorphous $(\text{Zr}_{1-x}\text{Er}_x)_3\text{Rh}$ alloys. Arrows show the position of the susceptibility maxima for $X = 0.15, 0.20$ and 0.225 . The magnetic transition is preempted by a superconducting transition at $T = 1.4$ K in the $X = 0.10$ sample.

Fig. 3. Magnetization as a function of applied field at several temperatures for amorphous $(\text{Zr}_{0.95}\text{Er}_{0.05})_3\text{Rh}$. Dashed line is a fit to data at $T = 4.65$ K and $H > 50$ kG as described in the text. Extrapolation to $H=0$ gives the spontaneous magnetization μ_s . Insert shows reciprocal initial susceptibility χ^{-1} as a function of temperature compared with a Curie-Weiss law fit. χ^{-1} is obtained by a least square fit to low field data as indicated by the solid lines in the $M(H)$ plot. Δ - $T = 3.3$ K; $\circ$ - $T = 4.7$ K; $+$ - $T = 8.7$ K; $\times$ - $T = 12.9$ K; $\blacktriangle$ - $T = 31.8$ K; $\bullet$ - $T = 50$ K; $\blacksquare$ - $T = 75$ K; $\square$ - $T = 100$ K.

magnetization exhibits two distinct regions of behavior. For $0 < H < 50$ kG, a rapid saturation of the magnetization is observed which gives over to a linearly increasing magnetization for $H > 50$ kG. This type of behavior has been previously observed in other amorphous alloys containing rare earths^{21,22} and can be explained in terms of a random local anisotropy which arises from crystal field effects at the Er site.^{22,23} By using a uniaxial anisotropy model, one can estimate the average crystal field splitting parameter. Following Fert and coworkers²¹ in their extension of the Zuckermann model²³, the high field magnetization can be expressed as

$$\langle \mu_z \rangle = - \frac{1}{2} g_J J \mu_B + \frac{g_J^2 \mu_B^2}{3D} (H_z + \lambda \langle \mu_z \rangle) \quad (1)$$

where $\langle \mu_z \rangle$ is the average polarization of Er in a field H_z applied along the z-direction and λ represents the mean molecular field. The anisotropy energy at site i is given by

$$H_i = D J_z^2 \quad (2)$$

where J_z is the z component of the total angular momentum and D is the crystal field parameter. At high fields, this becomes $\langle \mu_z \rangle = \alpha H_z$ where α is constant and D can be estimated from α ²¹ as

$$D = \frac{g_J^2 \mu_B^2}{3\alpha} \quad (3)$$

provided that $k_B \theta_p \ll D J^2$. We obtain $D/k_B \approx 5$ K from the least squares fit of the present data at high field ($H > 50$ kG) as shown in Fig. 3 so that

in the present case the above condition is fulfilled. The uniaxial model should be applicable to a Kramer's ion when D is positive and of the order found here. Linear extrapolation of the high field portion of the magnetization data (see Fig. 3) to $H = 0$ gives a spontaneous polarization of $\mu_S \approx 4 \mu_B$ per Er atom in excellent agreement with the value $\mu_S = 4.5 \mu_B$ obtained by Asomoza et al²¹ for amorphous Ni_3Er and also with the value of $\mu_S = \frac{1}{2} g_J \mu_B = 4.8 \mu_B$ expected for a Kramer's ion in the uniaxial model. We conclude that a random uniaxial crystal field model appropriately describes the Er environment in the present case.

Keller and Fulde¹⁰ have analyzed the role of crystal fields in modifying the theory of Abrikosov and Gor'kov²⁴(AG) for the depression of T_c by magnetic impurities. A Kramer's ion has a magnetic ground state which is at least doubly degenerate. In this case, elastic spin flip scattering of conduction electrons (or d-electrons) by the Er f-electrons will give rise to a finite pair breaking effect at all temperatures in contrast to the case of non-Kramer's ion with a non-magnetic ground state. Following Keller and Fulde, one expects a modification of the AG dependence of T_c on X resulting in a somewhat enhanced critical concentration for the disappearance of superconductivity. In Fig. 1, the AG theory is compared to the data by fitting to the initial rate of depression of T_c (resistively measured). The AG theory fits the data well for $X \leq 0.10$ if the critical concentration is taken to be $X_c = 0.13$ and $T_{c0} = 4.51$ K. Comparison with the theoretical curves of Keller and Fulde obtained by extending the AG theory to include the effect of a crystal field on a Kramer's ion suggests that the data for $X < 0.10$ can still be adequately described (including the effect of a crystal field). The peak observed in the T_c vs. X curve between $X = 0.11$

and $X = 0.14$ cannot however be accounted for using this extension of the AG theory. A correlation between this peak in $T_c(X)$ and the phase boundary of the magnetically ordered state, $T_M(X)$, is obviously suggested in Fig. 1.

Bennemann and Garland⁸ and Entel and Klose⁹, have analyzed the influence of magnetic ordering and the associated local molecular field on the pair-breaking behavior of interacting magnetic impurities. For a spin glass type of ordering one expects a rather small magnetic correlation length ξ_c . Poon and Durand²⁰ have estimated that $\xi_c \sim 20 \text{ \AA}$ for $(\text{La}_{1-x}\text{Gd}_x)_{80}\text{Au}_{20}$ alloys with X in the range of present interest. The zero temperature coherence length of the Zr_3Rh matrix can be estimated from H_{c2} data to be $\xi(0) \approx 50 \text{ \AA}$.²⁵ Thus $\xi(T) > \xi(0) \gg \xi_c$ in the present case and the molecular field below T_M should be zero when averaged over the typical dimension of a Cooper pair. Below T_M , spin-flip scattering, which gives rise to pair breaking, should become inelastic owing to the finite local molecular field at the Er site. Bennemann and Garland show that an anomalous dependence of $T_c(X)$ should result near $X = X_c$. Previous studies²⁵ of amorphous superconductors suggest strong spin-orbit scattering so that the above analysis should apply in the present case. In the limit of strong spin orbit scattering, they predict an enhanced critical concentration $X_c^{\text{BG}} = \left(\frac{J+1}{J}\right) X_c^{\text{AG}}$ where X_c^{AG} is the AG critical concentration and J is the total spin quantum number of the magnetic impurity. For Er^{3+} , $J = \frac{15}{2}$ so that $X_c^{\text{BG}} = 1.13 X_c^{\text{AG}}$. This is in approximate agreement with the experimentally observed $X_c^{\text{exp}} = 0.15 = 1.15 X_c^{\text{AG}}$. Although the Bennemann and Garland model predicts the enhancement of X_c , it cannot predict the observed peak in the $T_c(X)$ curve at $X = 0.12$. A model which accounts for short range magnetic correlations by Entel

and Klose predicts two superconducting regions in the $T_c(X)$ diagram separated by a non-superconducting region near the magnetic phase boundary. This model does predict a peak effect as observed in the present case.

Finally we turn to the behavior of the critical field $H_{c2}(X,T)$ to obtain additional insight into the nature of magnetic ordering. Data for $1.2 \text{ K} < T < 4.5 \text{ K}$ and $X = 0.0, 0.01, 0.05, 0.09$, and 0.10 are shown in Fig. 4. The behavior of $H_{c2}(X,T)$ can in the simplest approximation be described in terms of the average spin polarization of Er produced by spontaneous ordering or by an external field. Following Bennemann²⁶, we take

$$\begin{aligned} \vec{H}_{c2}(X,T,M=0) \\ = \vec{H}_{c2}(X,T,M) + \vec{I}(X,T,\vec{H})\mu_B^{-1} \end{aligned} \quad (4)$$

with

$$\vec{I} = \Gamma_{df} \vec{M} / g_J \mu_B \quad (5)$$

In these expressions, $\vec{I}$ is the exchange field at temperature T in applied field $\vec{H}$ and is related to the conduction (d-band) electron-4f electron exchange integral Γ_{df} . Γ_{df} can be estimated using the AG theory and the $T_c(X)$ data in the limit where $X \rightarrow 0$. Using a density of states $N(0) \approx 1.2$ (states/eV-atom) as suggested by specific heat data on a similar alloy $\text{Zr}_{70}\text{Pd}_{20}$ ²⁷ and the AG expression

$$(.75)^{-1} \left(\frac{dT_c}{dX} \right)_{X=0} = \frac{\pi^2 N(0) G \Gamma_{df}^2}{32k_B} \quad (6)$$

Fig. 4. The left hand side shows the reduced critical field $h(X, T) = [H_{c2}(X, T)/H_{c2}(0, 0)]$ as a function of reduced temperature (T/T_{c0}) where T_{c0} is the T_c of Zr_3Rh . The value of $H_{c2}(0, 0)$ has been replaced by $H_{c2}(0, T = 1.5 \text{ K}) = 92 \text{ kG}$ since $H_{c2}(0, 0)$ has not been measured. (see ref. 12) Data for several values of X are shown. The right hand side shows $h(X, T = 1.5 \text{ K})$ plotted as a function of (X/X_c^{AG}) . The dashed line shows $h_0(X)$ as defined by the knee in the curve. See text for further details.

gives $\Gamma_{df} = 0.06$ eV. Here, $G = (g_J - 1)^2 [J(J+1)]$ is the de Gennes factor. Note that the concentration X is multiplied by the factor $(0.75)^{-1}$ to give (dT_c/dX) in terms of the concentration of the whole alloy $(Zr_{1-X}Er_X)_{.75}Rh_{.25}$. A dramatic depression of H_{c2} can be seen in samples containing Er as compared to Zr_3Rh . This is consistent with a large Er contribution to $\vec{I}$.

Again following Bennemann et al²⁶, one can plot $h = [H_{c2}(X, T)/H_{c2}(X=0, T=0)]$ as a function of reduced concentration (X/X_C^{AG}) as shown in Fig. 4. For small Er concentrations and $T \ll T_c$, one has $\mu_B H_{c2} > k_B T$ and the Er spins should saturate along the applied field direction. As previously noted in analyzing the magnetization data, crystal field effects prevent saturation of the Er magnetization to the free ion value. Instead, one obtains $\mu_{Er} \approx 4\mu_B$ in agreement with the prediction $\mu_S \approx \frac{1}{2}g_J\mu_B$ of the uniaxial model. In any case, one expects $\vec{I} \propto \vec{H}$ for $T \ll T_c$ and X small so that h should decrease linearly with (X/X_C^{AG}) as illustrated in Fig. 4. As the Er concentration is increased, the magnetic ordering temperature $T_M (\theta_p)$ becomes comparable to T_c and the spontaneous local exchange field due to ordering must be taken into account. The sharp knee in $h(X/X_C^{AG})$ observed in Fig. 4 is clear evidence of the effect of magnetic ordering. From the form of this curve²⁶, one can deduce that the average magnetization $\langle \vec{\mu}_{Er} \rangle$ of Er in the magnetically ordered state is small. Average in this context refers to the magnetization averaged over Er spins within a typical volume of dimension ξ . We conclude that for $H = 0$, $\langle \vec{\mu}_{Er} \rangle_\xi \approx 0$ in the magnetically ordered state. This is the expected result for a spin glass type of ordering. The persistence of superconductivity for $X > X_C^{AG}$ has already been explained by the freezing out of spin flip scattering below the magnetic ordering

temperature. This analysis also required that $\langle \mu_{Er} \rangle_{\xi} \approx 0$ below $T_M(\theta_p)$. This is consistent with the H_{c2} analysis.

From the knee in the $h(X/X_C^{AG})$ curve one can estimate an effective ordering temperature $T_o(X)$. If we define $h_o(X)$ as

$$h_o(X) = \frac{k_B T_o(X)}{\mu_B H_{c2}(X=0, T=0)} \quad (7)$$

where $h_o(X)$ is experimentally determined by the knee in the $h(X/X_C^{AG})$ curve, then $T_o(X)$ is implicitly determined. From the data, one finds $T_o(X) \approx 16X(K)$. This effective ordering temperature compares to $\theta_p \approx 100X(K)$ from the high temperature Curie-Weiss fit and to $T_M(X) = 9.8X(K)$ from the susceptibility maxima data. The onset of magnetic ordering can also be estimated as the highest temperature, T_D , for which $\chi(T)$ deviates from a Curie-Weiss law (see Fig. 3). At $X = 0.05$, $T_D \approx 6K$ so that $T_D \approx 120X(K)$. Summarizing, one has $T_M \lesssim T_o \ll \theta_p \approx T_D$. It is concluded that the onset of magnetic ordering is gradual with lowering temperature. Magnetic clusters are formed as T is lowered below θ_p (or T_D). These clusters become configurationally frozen with respect to each other at a much lower temperature roughly given by T_M . Poon and Durand²⁰ have described magnetic ordering in $(La_{1-x}Gd_x)_{80}Au_{20}$ alloys using a magnetic cluster model of a concentrated spin glass. To the extent that crystal field effects can be neglected in the present case, the same model should be applicable.

In summary, we have shown that a magnetic ordering transition occurs in amorphous superconducting $(Zr_{1-x}Er_x)_3Rh$ alloys with $T_M \propto X$. Incipient ordering occurs at a temperature $\theta_p > T_M$ by formation of localized magnetic clusters which become configurationally frozen near T_M .

Superconductivity coexists with the magnetically ordered state over a broad range of compositions and is enhanced by the reduction of elastic exchange scattering (through magnetic ordering) for X near X_C^{AG} . The theories of Bennemann et al.⁸ and Entel and Klose⁹ qualitatively explain the behavior of $T_c(X)$. Measurements of the upper critical field $H_{c2}(X,T)$ give additional insight into the magnetic ordering. The behavior of $H_{c2}(X,T)$ can again be understood by a simple model proposed by Bennemann et al.²⁶ The model gives a means of estimating the magnetic ordering temperature from the $H_{c2}(X,T)$ data.

It is clear that additional studies of such alloys will be useful in clarifying the nature of magnetic order in amorphous alloys. Superconductivity is seen to be a sensitive microscopic probe which can be employed to study this phenomena. In particular, other alloys of the form $(Zr_{1-x}(R.E.)_x)_3Rh$ should be interesting. By suitable choice of the rare earth component, one can adjust X_C^{AG} to occur beyond the percolation threshold for ferromagnetic (or antiferromagnetic) ordering. Reentrance phenomena should be observed in this case. Such studies are presently in progress.

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