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Upper Critical Field of Superconducting Amorphous
Alloy $Zr_{85}Si_{15}$ *

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Synopsis

The upper critical field H_{C2} is measured for the amorphous superconductor $Zr_{85}Si_{15}$ ($T_c=2.70$ K, $\rho_n=270$ $\mu\Omega\cdot\text{cm}$) in the wide temperature range down to 50 mK. The conventional V-H recording in sweeping magnetic field and measurements of the flux flow resistivity defined by the I-V curve at constant field are done in order to clarify the resistive transition of the superconducting mixed-to-normal state in an amorphous superconductor with extreme softness in the flux pinning interaction. The results show the strong suppression of H_{C2} at low temperatures, which is quite different from the upward deviation from the theoretical values H_{C2}^* due to the orbital pair breaking effect in the dirty limit, reported for some amorphous alloys. Analysis with the Werthamer-Helfand-Hohenberg theory which takes into account the effects of the orbital pair breaking, spin paramagnetism and spin-orbit interaction gives $1.51 \leq \alpha \leq 1.77$ and $1.0 \leq \lambda_{SO} \leq 1.7$, where α is the impurity parameter and λ_{SO} the spin orbit interaction parameter. This is the first implication for the superconductivity in amorphous alloys that the spin paramagnetic effect plays an important role in determining the low temperature H_{C2} and the spin-orbit (spin-flip) scattering enhances H_{C2} moderately. The counterevidence is produced to the previous observation that the upward deviation anomaly ($H_{C2} > H_{C2}^*$) is an intrinsic property in amorphous metals with high resistivity.

Introduction

Recently there have been considerable interests in superconductivity of amorphous systems such as T-M and T-T alloys (T; transition metal, M; metalloid)⁽¹⁾. The most important findings character-

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izing their superconductivity are as follows.

(1) A highly disordered structure is responsible for short mean free path ($\sim \overset{\circ}{\text{Å}}$) leading extremely dirty type-II superconductivity with high GL parameter of about 100.

(2) A weak pinning interaction indicated by the low depinning current J_p is related with the fact that the amorphous phase is considered to be isotropic and homogeneous in a spatial scale over the Ginzburg-Landau coherence length ξ_{GL} of about 50-100 $\overset{\circ}{\text{Å}}$. Therefore, above J_p , the flux flow state is easily established ⁽²⁾.

(3) Relatively weak or intermediate coupling in electron-phonon interaction is considered to be realized ⁽¹⁾, which seems to be contrary to the single element amorphous superconductors such as Bi and Pb.

One of the puzzling problems lies in the upward deviation anomaly ⁽¹⁾⁽³⁾ of the upper critical field H_{C2} from H_{C2}^* the critical field expected theoretically on the basis of the orbital pair breaking effect in the dirty limit ($\xi_0 \gg \ell$, ξ_0 ; BCS coherence length, ℓ ; electron mean free path). This anomaly is observed in a lot of specimens but it is not well recognized to be intrinsic anomaly in an amorphous phase with high resistivity $\rho_n \geq 100 \mu\Omega \cdot \text{cm}$.

In this report we will describe the detailed experimental results for $H_{C2}(T)$ in $\text{Zr}_{85}\text{Si}_{15}$ ⁽⁴⁾ and show that our system exhibits a strong suppression of H_{C2} at low temperatures, contrary to the upward deviation anomaly.

Experimental

Amorphous ribbon specimens of about 1-2 mm width and 0.02-0.03 mm thickness were prepared by the liquid quenching technique using a modified single-roller melt spinning apparatus adapted to a levitation furnace. Identification of the as-quenched phase was done by the X-ray diffraction method and transmission electron microscopy.

The electrical resistance was measured by the four probe method with the electrical contacts by the high conductive silver paints. Above 1 K, specimens were immersed into the liquid He bath and the temperature was detected by the calibrated Ge thermometer and controlled within 10 mK over two hours by the manostat. Below 1 K, the ^3He - ^4He dilution refrigerator was used.

Definition of H_{C2}

We made two kinds of experiments under the isothermal condition; (1) the resistive transition in sweeping magnetic field at constant

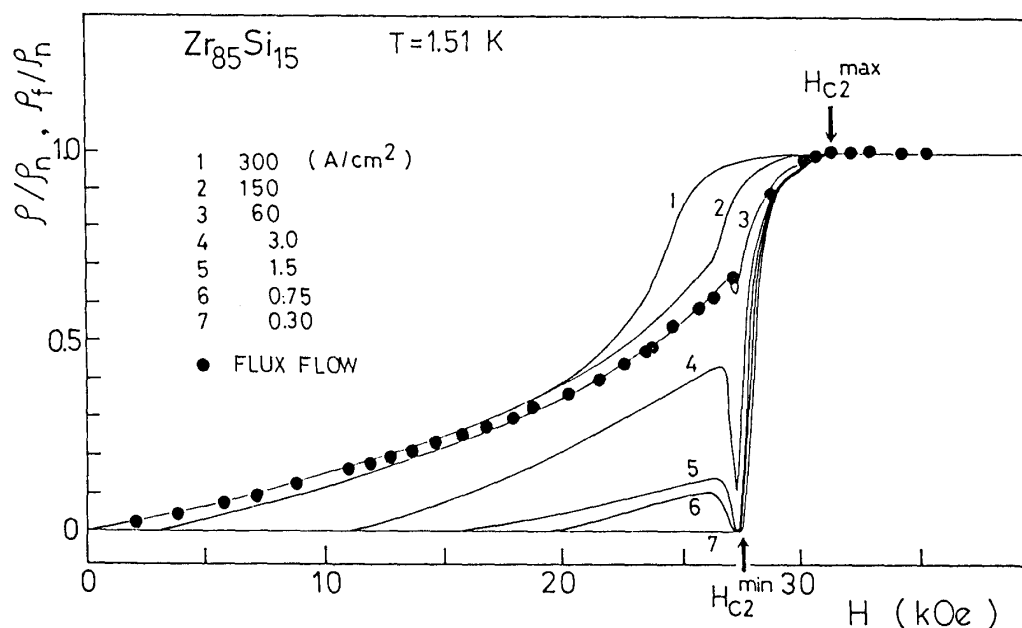


Fig. 1 Resistive transition curves under several currents and flux flow resistivity as a function of the magnetic field at $T = 1.51$ K for amorphous $Zr_{85}Si_{15}$.

dc currents ranging from 0.3 to 300 A/cm² and (2) the I-V curve measurement in constant magnetic fields that gives the flux flow resistivity $\rho_f = dV/dI$ and hence ρ_f/ρ_N as a function of H .

In Fig. 1 are shown the resistive transition curves under several currents by the solid lines and ρ_f/ρ_N -vs- H by the circles, at $T = 1.51$ K.⁽²⁾ In condition of low current (0.3 A/cm²) no resistance appears up to about 27 kOe. With increasing current, the resistive state becomes to be clearly seen in the wide field range. The transition dip near 27 kOe reflects a peak effect near H_{C2} , i.e., sharp increase of J_p .

In any type II superconductor the finite transition width is always observed and usual definition of H_{C2} is done in such conventional ways as the onset of resistivity and/or the midpoint. The transition width observed in the specimens will be described later. We define two critical fields, H_{C2}^{\max} and H_{C2}^{\min} , shown in the Figure, which means the fields giving the almost recovery of the resistivity ratio ($\rho/\rho_N, \rho_f/\rho_N \geq 0.99$) and the nucleation point of the resistivity in low current, respectively. These H_{C2}^{\max} and H_{C2}^{\min} indicate the upper and lower bound values in superconducting mixed-to-normal transitions. Below 1 K, where the flux flow resistivity measurement

were impossible due to heating, the resistive transition in the ac current ($0.5 \mu\text{A}_{\text{p-p}}$, 30 Hz) was measured and the upper critical fields were defined by the same criterion mentioned above.

Results and Discussions

The temperature dependence of H_{C2}^{max} and H_{C2}^{min} is shown in Fig. 2 (a) and (b), respectively. The dashed curve indicates the theoretical curve of H_{C2}^* . Both experimental values deviate downward from H_{C2}^* , showing the strong suppression at low temperatures. In Table I experimental values are tabulated. T_{C0} is the midpoint temperature in the transition curve in absence of the field and $T_{C(H \rightarrow 0)}$ the temperature obtained by extrapolating H_{C2} curve to $H = 0$. The discrepancy between two $T_{C(H \rightarrow 0)}$ values in cases of H_{C2}^{max} and H_{C2}^{min} is reasonably explained, taken into account the fact that the full transition width in absence of the field is about 0.1 K.⁽⁴⁾

The present results are compared with the theory extended by Werthamer, Helfand and Hohenberg (WHH)⁽⁵⁾ to include the effects of Pauli spin paramagnetism⁽⁶⁾ and spin-orbit interaction. The H_{C2} values for isotropic type II superconductors in the dirty limit are numerically calculated at an arbitrary temperature using the following equation with digamma function Ψ

$$\ln \frac{1}{t} = \left(\frac{1}{2} + \frac{i\lambda_{SO}}{4\gamma} \right) \Psi \left(\frac{1}{2} + \frac{\bar{h} + (1/2)\lambda_{SO} + i\gamma}{2t} \right) + \left(\frac{1}{2} - \frac{i\lambda_{SO}}{4\gamma} \right) \Psi \left(\frac{1}{2} + \frac{\bar{h} + (1/2)\lambda_{SO} - i\gamma}{2t} \right) - \Psi \left(\frac{1}{2} \right), \quad (1)$$

where

$$t = \frac{T}{T_C}, \quad (2)$$

Table I Experimental values for amorphous $\text{Zr}_{85}\text{Si}_{15}$ (3)

| ρ_n ($\mu\Omega \cdot \text{cm}$) | T_{C0} (K) | $T_{C(H \rightarrow 0)}$ (K) | $-dH_{C2}/dT$ (kOe/K) T_C | $H_{C2}(0)$ (kOe) |
|---|-----------------|---------------------------------|--------------------------------|----------------------|
| 270 | 2.70 | * 2.73 | 33.7 | 47 |
| | | ** 2.64 | 28.8 | 43 |

* For H_{C2}^{max}
** For H_{C2}^{min}

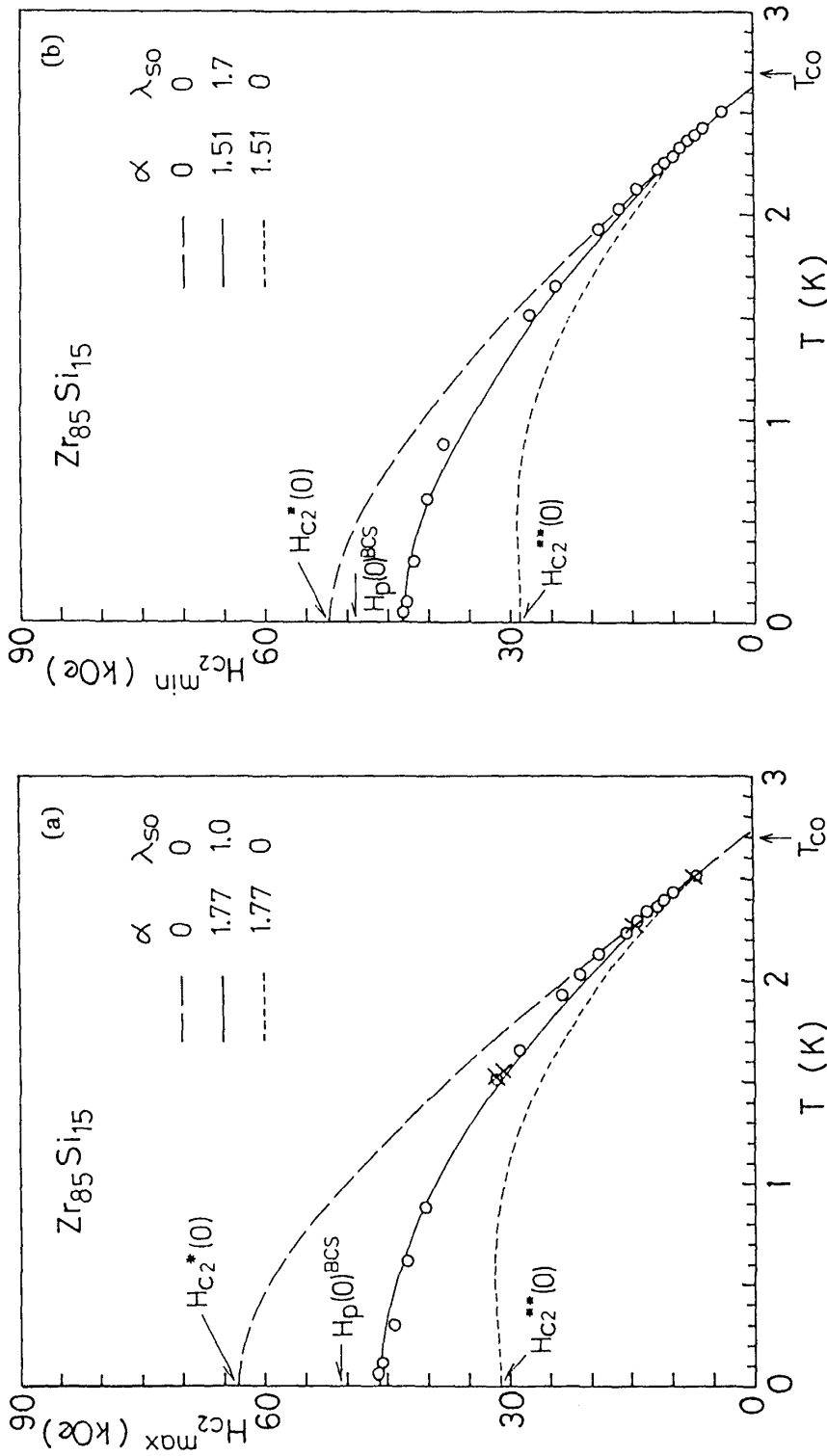


Fig. 2 Upper critical field H_{c2} as a function of temperature. (a); H_{c2}^{max} and (b) H_{c2}^{min} . The circle and cross indicate the experimental values obtained by V-H recording (ρ/ρ_n vs H) and flux flow resistivity (ρ_f/ρ_n vs H). The theoretical (WHH) curves are for only orbital pair breaking effect without spin paramagnetism, shown by dashed lines, for spin paramagnetism without spin-orbit effect, shown by dotted lines, and for spin paramagnetism as well as a best-fit-adjusted spin-orbit interaction parameter, shown by solid lines.

$$\bar{h} = \frac{2eH v_F^2 \tau_{tr}}{c 6\pi k_B T_C} , \quad (3)$$

$$\gamma = \left[(\alpha \bar{h})^2 - (\lambda_{SO}/2)^2 \right]^{1/2} , \quad (4)$$

$$\alpha = \frac{3\bar{h}}{2m v_F^2 \tau_{tr}} \quad (5)$$

and

$$\lambda_{SO} = \frac{\bar{h}}{3\pi k_B T_C \tau_{SO}} . \quad (6)$$

The dimensionless quantities α and λ_{SO} are so-called impurity (Maki) parameter and spin-orbit interaction parameter, respectively.

If one assumes $\alpha = \lambda_{SO} = 0$, i.e., in absence of the spin paramagnetic effect and the spin-orbit interaction, one gets following simple expression for H_{C2} due to orbital pair breaking.

$$\ln \frac{1}{t} = \Psi \left(\frac{1}{2} + \frac{\bar{h}}{2t} \right) - \Psi \left(\frac{1}{2} \right) , \quad (7)$$

which gives $H_{C2} = H_{C2}^*(T)$ curve shown by the dashed line in Fig. 2.

The parameter α is uniquely defined by the experimental value of $(-dH_{C2}/dT)_{T_C}$ (in kOe) as follows.

$$\alpha = \sqrt{2} H_{C2}^*(0) / H_p(0)^{BCS} = 5.276 \times 10^{-2} \cdot (-dH_{C2}/dT)_{T_C} \quad (8)$$

because

$$H_{C2}^*(0) = 0.69 \cdot (-dH_{C2}/dT)_{T_C} \quad (9)$$

and

$$H_p(0)^{BCS} = \Delta_0 / \sqrt{2} \mu_B = 18.57 T_C \quad (10)$$

where Δ_0 is the energy gap at $T = 0$ and μ_B the Bohr magneton.

For comparison, the values of λ_{SO} are adjusted under the fixed values of T_C and α so that the experimental values at low temperatures (50, 100 mK) are fit with the corresponding theoretical values. It is found that the temperature dependence is well reproduced by the theoretical curve derived by setting $\alpha = 1.77$ and $\lambda_{SO} = 1.0$ for H_{C2}^{\max} and $\alpha = 1.51$ and $\lambda_{SO} = 1.7$ for H_{C2}^{\min} , as shown by the solid line in Fig. 2 (a) and (b). The dotted curve shows $H_{C2}^{**}(T)$ determined by setting $\lambda_{SO} = 0$, in absence of a spin-orbit scattering effect. In Table II are

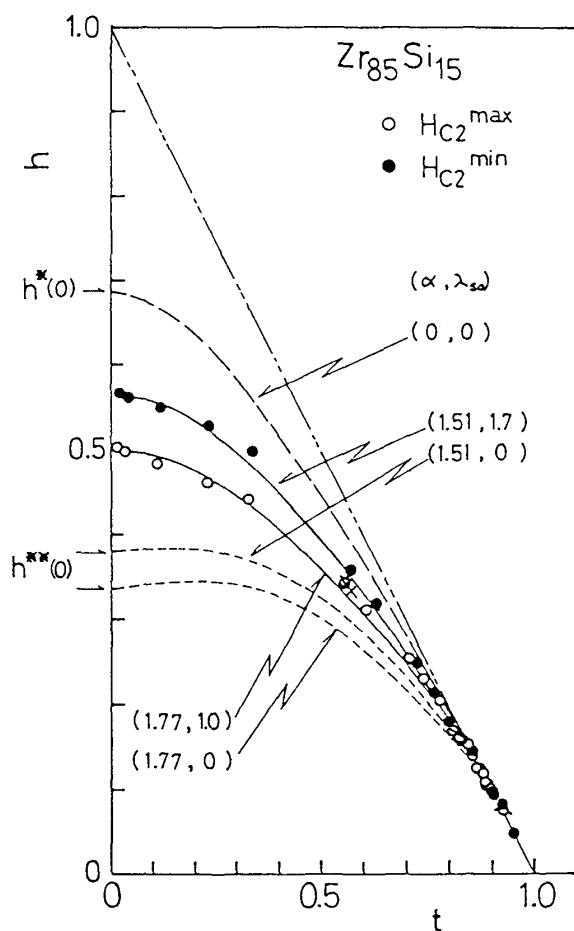


Fig. 3 A plot of the normalized critical field h as a function of the reduced temperature t . The content is the same as in Fig. 2.

Table II Quantities derived by the analysis with WHH theory.

| | $H_{c2}^*(0)$ (kOe) | $H_{c2}^{**}(0)$ (kOe) | $H_p(0)^{BCS}$ (kOe) | α | λ_{50} |
|----|------------------------|---------------------------|-------------------------|----------|----------------|
| * | 63.4 | 31.2 | 50.7 | 1.77 | 1.0 |
| ** | 52.3 | 28.9 | 49.0 | 1.51 | 1.7 |

* For H_{c2}^{\max}

** For H_{c2}^{\min}

are tabulated the quantities derived by the analysis based on the WHH theory. In Fig.3 is shown the dimensionless field h vs temperature t , converted from Fig. 2 (a) and (b) using the definition,

$$h = \frac{H_{C2}(T)}{(-dH_{C2}/dT)_{T_C} \cdot T_C} \quad (11)$$

We discuss the validity of the theory in applying it to our disordered system. Using Eq. (6) we obtain $\tau_{SO} = 2 \times 10^{-13}$ sec much longer than transport relaxation time $\tau_{tr} = 10^{-15} - 10^{-16}$ sec derived by assuming Fermi velocity $v_F = 10^7 - 10^8$ cm/sec for the specimen having $\rho_n = 270 \mu\Omega \cdot \text{cm}$. This result well satisfies the required assumption in the theory that the spin-flip scattering should be infrequent in comparison with non-spin-flip scattering, i.e., $\tau_{SO}^{-1} \ll \tau_{tr}^{-1}$. The second assumption neglecting the strong electron-phonon coupling may have no decisive role in determining $H_{C2}(T)$. The electron-phonon coupling parameter $\lambda_{ep} = 0.6$ is obtained using the McMillan formula with $\mu^* = 0.15$ and $\Theta_D = 220$ K, the latter derived by the Young's modulus measurement⁽⁷⁾. This implies that our system is a weak or intermediate coupling superconductor, which has been recognized recently in many systems of transition-metal based amorphous alloys⁽¹⁾.

Through these considerations we suggest that the spin paramagnetic effect may play an important role in determining the low temperature H_{C2} and spin-orbit (spin-flip) scattering enhances H_{C2} moderately. This is the first implication for the superconductivity in amorphous alloys and produces the counterevidence to the previous observation⁽¹⁾⁽³⁾ that upward deviation anomaly, $H_{C2} > H_{C2}^*$, may be a unique property in amorphous phase with high resistivity $\rho_n > 100 \mu\Omega \cdot \text{cm}$.

One possible explanation for this anomaly was proposed on the basis of the phenomenological model⁽⁸⁾ treating the inhomogeneity effect to H_{C2} as a spatial variation of the diffusion constant. The numerical results reveal the trend of upward deviation of H_{C2} from H_{C2}^* with increasing a degree of distribution. The spatial variation of critical values such as T_C , H_{C2} and J_p should be considered responsible for the inhomogeneity.

The problem related with inhomogeneity is very difficult to be dealt with, but one may think two types of inhomogeneity; (1) metallurgical one causing the spatial variation in the atomic concentration, local strain fields etc. and (2) intrinsic one due to static fluctuation of the short range order in topological and chemical bonding nature.

We have recognized⁽²⁾ the importance of the strength in depin-

ning current which is one of the most sensitive measures⁽⁹⁾ to the inhomogeneity in real amorphous phase through the pinning interaction between fluxoids and inhomogeneous regions. Our specimen has $J_p = 10^0$ A/cm² at $H/H_{C2} = 0.5$ which are considered low compared with some specimens showing upward deviation anomaly. Therefore it is reasonably expected that the upward deviation anomaly may be strongly related with the degree of inhomogeneity influencing J_p drastically.

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