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Fluctuation superconductivity at high magnetic fields*

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Apparent superconductive fluctuation conductivity ("paraconductivity") $\Delta\sigma_f$ has been observed in very "dirty" (short electron-mean-free-path), bulk, type-II superconductors in applied magnetic fields H up to twice the zero-temperature upper critical field H_{c20}. Peaks observed in the isomagnetic field paraconductivity as a function of temperature at $H > H_{c20}$ are attributed to the suppression of paraconductivity as T approaches either zero or values high in comparison with the zero-H transition temperature T_c. In the (H-T) region well beyond the upper-critical-field curve $H_{c2}(T)$, the experimentally derived $\Delta\sigma_f(H, T)$ is smaller, less dependent on the orientation of H with respect to the measuring current density J, and decreases more rapidly with H than suggested by current theory. As H is increased isothermally in the 80–140-kG region, the positive magnetoresistance associated with the H quenching of paraconductivity gives way to a small negative magnetoresistance which is probably not explicable on an ordinary static-localized-magnetic-moment basis.

There has been recent interest in the study of the effects associated with thermodynamic fluctuations of the superconductive order parameter, especially those effects which occur[1–3] at temperatures T and/or applied magnetic fields H well outside the (H-T) realm usually associated with equilibrium superconductivity. In this regime, standard theory often breaks down and new insight into superconductive interactions may be attained. In the present article[4] we report on the investigation of superconductive fluctuation conductivity ("paraconductivity") in very "dirty," bulk, type-II superconductors in applied magnetic fields H up to 140 kG. The present 2.5-fold increase in the H-field capability over that of previous work[3] has allowed the observation of near H quenching of the paraconductivity and the resultant determination of a reasonably accurate normal-state conductivity $\sigma_n(H, T)$. The more accurate base line $\sigma_n(H, T)$ has then allowed a better determination of the absolute magnitude of the paraconductivity $\Delta\sigma_f(H, T)=\sigma(H, T) - \sigma_n(H, T)$ and thus a better comparison of data with theory than was previously[3] possible, especially well above the upper-critical-field curve $H_{c2}(T)$ where $\Delta\sigma_f(T)$ is small. In addition, we have observed (apparently for the first time in bulk, type-II superconductors): (i) paraconductivity up to about twice the zero-temperature upper critical field H_{c20} (the paraconductivity does not appear to be correctly described by current theory); (ii) peaks in the isomagnetic-field $\Delta\sigma_f(T)$ curves at $H> H_{c20}$, which we attribute to the suppression of paraconductivity as T approaches either zero or values high in relation to the zero-H transition temperature T_c; (iii) a small negative magnetoresistance in the 100–140-kG range, which is probably not due to the presence of ordinary static localized magnetic moments.

Figure 1 shows magnetoresistive characteristics as measured for a typical,[5] very dirty,[6] paramagnetically limited,[5,7] superconducting alloy $Ti_{92}Ru_8$ in the (H-T) region beyond both the upper- and surface-[5] critical-field curves shown in Fig. 2. Magnetoresistive curves very similar to those of Fig. 1 have been measured for $Ti_{84}Mo_{16}$, $Ti_{92}Fe_8$, $Ti_{92}Os_8$, $Ti_{75}V_{25}$, $Ti_{86}Mn_{14}$, and $V_{60}Ti_{30}Cr_{10}$ and will be reported in a subsequent paper. The magnetoresistance measurements were made with a precision of about 3×10^{-5}, employing a standard dc 4-lead technique,[5] with an H-field homogeneity of one part in 10^3 over the specimen volume. The positive, saturating magnetoresistive curves of Fig. 1 are attributed[3] to the H quenching of superconductive fluctuations ("magnetoparaconductivity") rather than to ordinary normal-state magnetoresistance. This interpretation is consistent with (a) the high electrical resistivities of the present alloys ($\rho> 10^{-4}$ Ωcm) which suggest, on the basis of Kohler's-rule arguments,[3] negligible ordinary magnetoresistance, and (b) the nearly flat magnetoresistive characteristics observed in liquid neon at 27 K ($\approx 8T_c$), where near absence of fluctuations would be expected.[2,3]

The field $H_f(T)$, indicated in Fig. 1, is defined as the field at which the slope of an isothermal magnetoresistive curve is zero, and is regarded merely as an experimentally convenient measure of a least upper bound for the existence of detectable paraconductivity under the present experimental conditions. It should be emphasized that H_f serves only as a rough measure of the highest H at which paraconductivity could be investigated in the present work and has little or no analytic significance for fluctuation theory. Plots of $H_f(T)$, such as Fig. 2, yield $H_f(T_c)/H_{c20}=2.0\pm0.2$ for $Ti_{92}Ru_8$, $Ti_{84}Mo_{16}$,[8] and $Ti_{92}Fe_8$. As far as we are aware, the present H_f values represent the highest fields, in relation to H_{c20}, at which effects associated with superconducting fluctuations have thus far been observed in type-II superconductors.

A prominent feature of the magnetoresistive curves of Fig. 1 is their progressive sharpening with decrease of temperature, along with the associated decrease of H_f shown in Fig. 2. We attribute these effects [and related peaks in $\Delta\sigma_f$ $(T, H > H_{c20})$ shown in Fig. 3] to the decrease in fluctuation-forming thermal energy kT as T decreases. The crossover in the curves of Fig. 1 at $H \approx 60$ kG is related both to the sharpening of the curves and to the anomalous increase in the resistivity [higher $(V - V_s)/V_s$] at the highest H as temperature is reduced. The negative slope of the curves (negative magnetoresistance) at the highest H has apparently not been previously observed in alloys of the pres-

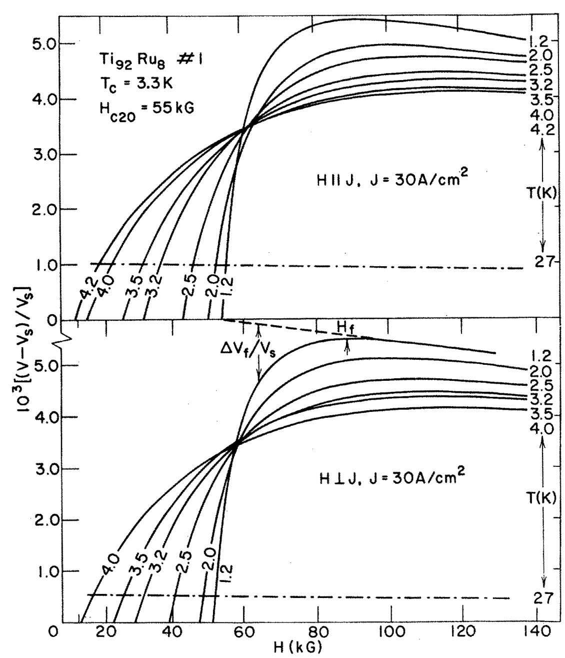

FIG. 1. Magnetoresistive curves for $Ti_{92}Ru_8$ as discussed in the text. V is the resistive voltage and V_s is a constant nulling voltage applied with a six-dial μV potentiometer [$V_s(H \parallel J) = 4392.50$ μV, $V_s(H \perp J) = 4110.00$ μV]. V_s is chosen so as to offset most of the resistive voltage and thus allow a large amplification of the off-balance signal with consequent increase in the precision of the measurement. The normalized difference voltage $(V - V_s)/V_s$ vs H is traced from X-Y recorder plots of $(V - V_s)$ vs V_{mr}, where V_{mr} is a voltage generated by a magnetoresistive field sensor. The lower plot indicates the extrapolation procedure for deriving $\Delta V_f/V_s$ and thus $\Delta\sigma_f/\sigma_n = (\Delta V_f/V_s)(V_s/V)$ as discussed in Ref. 19. The various isothermal curves were measured at different temperatures which are indicated along the right-hand side of the figure. The curves shown for $T = 27$ K have been elevated by about 2×10^{-3}.

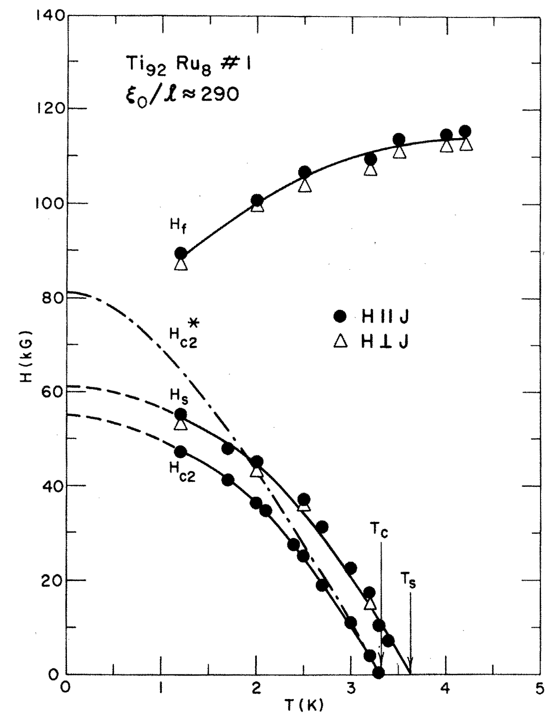

FIG. 2. Resistively determined upper critical field $H_{c2}(T)$ and surface critical field $H_s(T)$ for $Ti_{92}Ru_8$. The theoretical, nonparamagnetically limited, upper-critical-field curve $H_{c2}^*(T)$ is in accord with Ref. 7 and the presently measured $(dH_{c2}/dT)_{T_c}$ (see Ref. 6). H_f is merely a rough measure of the highest H at which paraconductivity could be investigated in the present work, as discussed in the text.

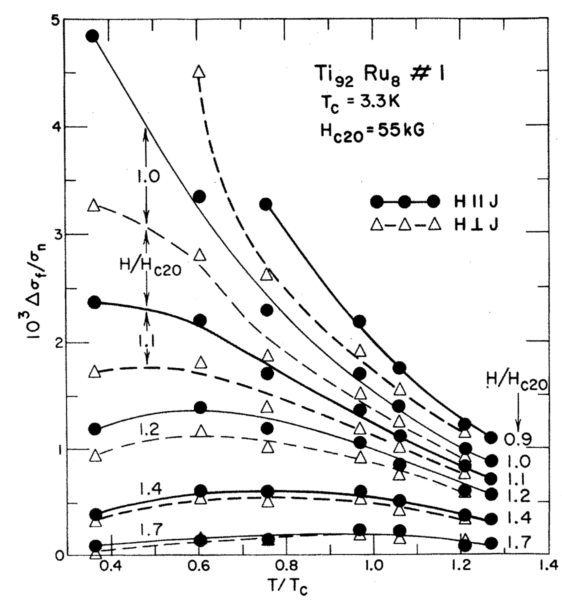

FIG. 3. Experimental normalized paraconductivity $\Delta\sigma_f(T/T_c)/\sigma_n$ for various constant normalized measuring fields H/H_{c20}.

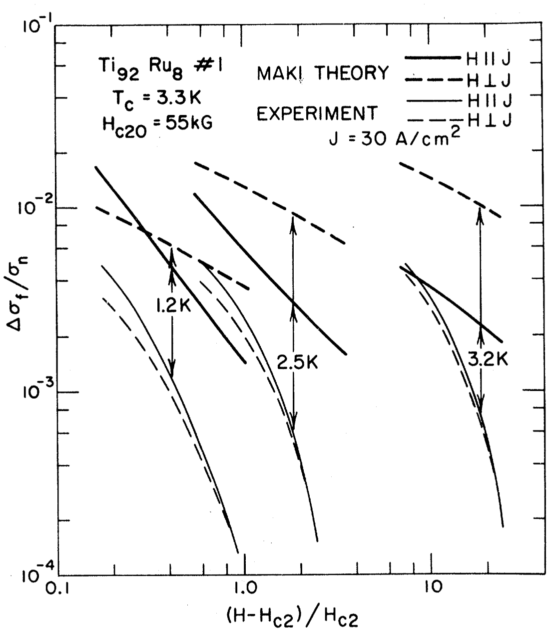

FIG. 4. Experimental normalized paraconductivity $\Delta\sigma_f/\sigma_n$ for various constant measuring temperatures, plotted as a function of $(H-H_{c2})/H_{c2}$. The theoretical curves represent the Maki expressions (Ref. 22), based on the Aslamazov-Larkin diagram, corrected for the paramagnetic limitation as discussed in the text. The theoretical $\Delta\sigma_f$ values have been divided by the measured σ_n (4.2 K, 140 kG) = 7.0×10^3 Ω cm^{-1}. In order to avoid complications due to the sheath, the experimental $\Delta\sigma_f/\sigma_n$ curves are derived from data taken only at $H>H_s(T)$, where $H_s(T)$ is the apparent measured surface critical field shown in Fig. 2.

ent type.

The weak, high-H, negative magnetoresistance shown in Fig. 1 is especially noteworthy because standard localized-magnetic-moment behavior[9] (which is usually associated[9] with negative magnetoresistance in metallic systems) has never, to our knowledge, been observed in bcc Ti-base alloys, even though calorimetric,[10] susceptibility,[11] magnetization,[5] and electrical-resistance[3,5,12–15] measurements have been made on many different specimens, some containing more than 5-at.% Cr, Mn, or Fe. We suggest[15] that the presently observed negative $(\partial\rho/\partial T)_H$ and negative $(\partial\rho/\partial H)_T$ at $1.2\leq T\leq 27$ K and $0\leq H\leq 140$ kG may both result from conduction-electron interaction with highly compensated or very rapidly fluctuating localized spins,[16] as might also account for the characteristic[17] high ρ (hence high H_{c20}) and negative[18] $(\partial\rho/\partial T)_{H=0}$ at $\approx 10 < T < 300$ K.

Figure 3 shows experimentally derived normalized paraconductivity $\Delta\sigma_f(T)/\sigma_n = (\sigma-\sigma_n)/\sigma_n$ at various normalized measuring fields H/H_{c20}. Here $\sigma_n(H, T)$ is the normal-state conductivity and $\Delta\sigma_f(T)/\sigma_n$ is derived from the isothermal curves of Fig. 1 by the linear extrapolation procedure indicated in that figure.[19] The maxima in the normalized paraconductivity curves of Fig. 3 (Ref. 20) are not unreasonable: above H_{c20} isofield superconductive fluctuations should die out as T approaches either zero or values high in relation to T_c.[2,3] Paraconductive peaks of this nature were apparently first predicted by Thompson[21] for thin films and are implicit in the three-dimensional expressions for $\Delta\sigma_f(T, J\parallel H)$ derived by Maki[22] and by Usadel[23] and based on the Aslamazov-Larkin (AL) diagram.[24]

Figure 4 shows normalized experimental paraconductivity curves $\Delta\sigma_f/\sigma_n$ plotted as functions of $(H-H_{c2})/H_{c2}$. Curves for $J=3$ A/cm^2 are nearly identical to the $J=30$ A/cm^2 curves shown in Fig. 4. The rather minimal anisotropy[25] for $J\parallel H$ and $J\perp H$, and the rapid decrease of paraconductivity with increase of H are notable. The theoretical curves of Fig. 4 represent the $\Delta\sigma_f$ expressions of Maki[22] based on the AL diagram but corrected for the paramagnetic limitation in a manner consistent with that suggested by Fulde and Maki.[26] The inclusion of (a) the Maki term,[27] (b) higher Landau-level contributions to the AL term,[23,28] and (c) other possible terms[23,27] which are nondivergent at H_{c2}, would only serve to elevate the theoretical curves and thus increase the discrepancy with experiment. Thus it appears that current microscopic theory[22,23,26,27] does not properly describe the present observations of paraconductivity well outside the upper-critical-field curve, where the effects of high-energy, very-short-wavelength fluctuations should become especially important.[2]

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[6]From the measured helium-temperature normal-state resistivity $\rho_n = 1.44\times10^{-4}\Omega$ cm, the resistively measured upper-critical-field slope at the zero-field transition temperature T_c, $(dH_{c2}/dT)_{T_c} = -35$ kG/K, and standard formulas and assumptions (Ref. 5), we deduce the "dirtiness" parameter $\xi_0/l\approx 290$, where ξ_0 is the BCS coherence distance and l is the electron mean free path. Also of interest here are $H^*_{c20} = 0.69T_c\,(-dH_{c2}/dT)_{T_c} = 81$ kG, $\xi(0) = (c\hbar/2eH^*_{c20})^{1/2} = 64$ Å, and $D = \frac{1}{3}V_F l = \varphi_0\Delta_{00}(2\hbar\pi H^*_{c20})^{-1}$

$=0.30\ \mathrm{cm}^2\ \mathrm{sec}^{-1}$, where H^*_{c20} is the theoretical zero-T nonparamagnetically limited upper critical field (Ref. 7), $\xi(0)$ is the zero-T Ginzburg-Landau coherence distance, D is the electronic diffusion constant, V_F is the Fermi velocity, φ_0 is the flux quantum, and Δ_{00} is the zero-T BCS half-energy gap.

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[8]Since both H_{c20} and T_c peak at $Ti_{84}Mo_{16}$ in the Ti_xMo_{100-x} system, the magnetoresistive and paraconductive characteristics in $Ti_{84}Mo_{16}$ (similar to those of Figs. 1–4) cannot be ascribed to H quenching of superconductivity in localized high-upper-critical-field regions.

[9]For reviews, see, e.g., C. Rizzuto, Rep. Prog. Phys. 37, 147 (1974) and M. D. Daybell, in *Magnetism V*, edited by H. Suhl (Academic, New York, 1973), p. 121.

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[15]J. W. Lue, A. G. Montgomery, and R. R. Hake, in *Proceedings of the 20th Annual* Conference on *Magnetism and Magnetic* Materials, San Francisco, 1974 (to be published). It is suggested that bcc Ti-base alloys are characterized by Kondo or spin-fluctuation temperatures $\theta > 300$ K, corresponding to Kondo or spin-fluctuation fields $\mathcal{H} \equiv k_B\theta/\mu_B > 4.6$ MG.

[16]Spin-disorder scattering was suggested in Ref. 12 as a mechanism for negative $(\partial\rho/\partial T)_{H=0}$ in bcc Ti-Mo alloys. Localized-spin fluctuations have previously been invoked in the case of bcc Ti-V alloys: (a) by A. F. Prekul, V. A. Rassokhin, and N. V. Volkenshtein, Zh. Eksp. Teor. Fiz. Pis'ma Red. 17, 354 (1973) [JETP Lett. 17, 252 (1973)], and V. A. Rassokhin, N. V. Volkenshtein, A. P. Romanov, and A. F. Prekul, Zh. Eksp. Teor. Fiz. 66, 348 (1974) [Sov. Phys.-JETP (to be published)] to account for negative $(\partial\rho/\partial T)_{H=0}$; and (b) by K. H. Bennemann and J. W. Garland, Int. J. Magn. 1, 97 (1971) and AIP Conf. Proc. 4, 103 (1972), to account for the magnitude of T_c.

[17]See, e.g., Refs. 12, 14, and 15.

[18]The recent survey of J. H. Mooij, Phys. Status Solidi A 17, 521 (1973) shows that for all disordered transition-metal alloys with low-temperature coefficients of resistance (considering data at ≈ 300–350 K) (a) a negative $(\partial\rho/\partial T)_{H=0}$ is found in nearly all materials with $\rho > 1.3\times10^{-4}\ \Omega\,\mathrm{cm}$, and (b) $\rho^{-1}(\partial\rho/\partial T)_{H=0}$ is roughly proportional to ρ. Both (a) and (b) are consistent with the high Kondo or spin-fluctuation temperature model suggested in Ref. 15.

[19]It is assumed that for each isothermal magnetoresistance curve the linear magnetoresistance at $H > H_f$ is a good approximation to the normal-state curve $(V_n - V_s)/V_s$, and that the normal-state magnetoresistance is linear over a wide-field range. (The latter assumption is supported by the linearity of the 27-K data shown in Fig. 1.) Thus, for example, the dashed-line linear extrapolation shown in the bottom portion of Fig. 1, near the curve difference label "$\Delta V_f/V_s$", is taken to represent $(V_n - V_s)/V_s$ for $H < H_f$ at $T = 1.2$ K, $H \perp J$. The labeled curve difference is then $\Delta V_f/V_s = [(V_n - V_s)/V_s] - [(V - V_s)/V_s] = (V_n - V)/V_s$. Multiplication of this curve difference by V_s/V (where V is, of course, a function of H and T) yields $[(V_n - V)/V_s](V_s/V) = (V_n - V)/V = (\sigma_n^{-1} - \sigma^{-1})/\sigma^{-1} = (\sigma - \sigma_n)/\sigma_n \equiv \Delta\sigma_f/\sigma_n$, since for constant measuring current density J and potential-lead spacing d, $V_n = \sigma_n^{-1}Jd$ and $V = \sigma^{-1}Jd$.

[20]Related *minima* in *resistance* versus temperature curves of thin Al films have been observed by P. M. Tedrow and R. Meservy, Phys. Rev. B 8, 5098 (1973) and K. Aoi, R. Meservy, and P. M. Tedrow, Phys. Rev. B 9, 875 (1974) (see also Ref. 26). In that case the effects appear to be related to the onset of first-order upper-critical-field transitons, whereas here neither resistive measurements near H_{c2} nor direct magnetization measurements (Ref. 5) yield any evidence for first-order transitons. Tedrow *et al.* also observed a negative temperature coefficient of resistance and a linear negative magnetoresistance in Al films at high H.

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[24]L. G. Aslamazov and A. I. Larkin, Fiz. Tverd. Tela 10, 1104 (1968). [Sov. Phys.-Solid State 10, 875 (1968)].

[25]Near H_{c2} the theoretically predicted (Refs. 22, 23, and 28) $\Delta\sigma_f(H \parallel J) > \Delta\sigma_f(H \perp J)$ and related H-induced one-dimensionality has apparently been observed: R. R. Hake, in *Low Temperature Physics-LT13*, edited by K. D. Timmerhaus, W. J. O'Sullivan, and E. F. Hammel (Plenum, New York-London, 1974), p. 638; R. F. Hassing, R. R. Hake, and L. J. Barnes, Phys. Rev. Lett. 30, 6 (1973).

[26]P. Fulde and K. Maki, Z. Phys. 238, 233 (1970). In the present case $\Delta\sigma_f$ is calculated from Eqs. (6), (11), and (16) of Ref. 22 [the right-hand side of Eq. (11) was multiplied by a factor $\frac{1}{4}H$ so as to make it consistent with Eq. (31) of Ref. 26] and then plotted as in Fig. 4 versus $(H - H_{c2})/H_{c2}$. Here the resistively-measured paramagnetically-limited upper-critical-field H_{c2} merely replaces the theoretical non-paramagnetically-limited H^*_{c2}, the field at which the theoretical $\Delta\sigma_f$ would diverge in the absence of the paramagnetic limitation. This method of treating the paramagnetic limitation is consistent with the Fulde-Maki prescription for $H \gtrsim H_{c2}$ in that H^*_{c2} is replaced by H_{c2}.

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