4011 (1967).

[11] V. R. Parameswaran and J. Weertman, Scripta Met. 3, 477 (1969).

[12] K. Marukawa, J. Phys. Soc. Japan 22, 499 (1967); see also T. Suzuki, in *Dislocation Dynamics,* edited by A. R. Rosenfield, G. T. Hahn, A. L. Bement, and R. I. Jaffee (McGraw-Hill, New York, 1968), p. 551.

[13] R. M. Fisher and J. S. Lally, Can. J. Phys. 45, 1147 (1967).

[14] G. P. Huffman and N. Louat, Phys. Rev. 176, 773 (1968).

[15] G. P. Huffman and N. Louat, in Proceedings of the Conference on Fundamental Aspects of Dislocation Theory, Gaithersburg, Md., April 1969 (unpublished).

[16] I. A. Privorotskii, Zh. Eksperim. i Teor. Fiz. 43, 1331 (1962) [Soviet Phys. JETP 16, 945 (1963)].

[17] P. G. De Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966), Chap. 4.

[18] J. R. Schrieffer, *Theory of Superconductivity* (Benjamin, New York, 1964), Chaps. 2 and 3.

[19] T. Holstein, in an appendix to Ref. (4). Recently, the present authors have given a theory of dislocation drag in the normal state based on the Boltzmann-equation formalism [Ref. (10)]. The results indicated that the temperature-independent scattering result of Holstein is probably correct for close-packed metals. However, for bcc metals the electronic drag was found to be proportional to the electrical conductivity. As it has not yet been possible to derive this latter result from a scattering theory, we will take the dislocation drag in the normal state as that given by Eq. (17) in this paper. At any rate, one might hope that the ratio of the drag in the superconducting state to that in the normal state will not be greatly changed, whatever the form of the normal state drag coefficient. We hope to treat this question in more detail in a future paper.

[20] The results for α_1 and α_2, which are simply the attenuation ratios of a high-frequency phonon, are quite interesting in themselves and will be given in a separate paper. The temperature dependence of the energy gap in all these calculations has been taken as that given by B. Mühlschlegel, Z. Physik 155, 313 (1959).

[21] A. Granato and K. Lücke, J. Appl. Phys. 27, 583 (1956).

EVIDENCE FOR FLUCTUATION EFFECTS ABOVE T_c IN ISOTOPICALLY AND METALLURGICALLY PURE BULK TYPE-I SUPERCONDUCTORS*

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Magnetic measurements of superconducting transitions of very pure isotopically separated bulk samples of Ga and Zn reveal a large "paraconductive" effect. This is contrary to the prediction for such systems by Aslamazov and Larkin based on corrections to first order in the fluctuations in the system. It is suggested that this discrepancy may be explained by increased electron pair lifetimes due to the absence of isotopic and other scattering of phonons allowing higher order processes to be significant.

According to the theory of Aslamazov and Larkin (AL),[1] the electrical conductivity of a superconducting material in the normal state increases as the temperature approaches T_c because of fluctuations in the system (paraconductivity). This effect can be large for $T-T_c \sim 10^{-3}$ K in thin films and whiskers, but for pure bulk type-I superconductors, appreciable paraconductivity should be confined to the presently unobservable temperature range $T-T_c \sim 10^{-15}$ K. The theory predicts both a magnitude and a temperature dependence for the effect. Measurements on a variety of thin films[2] have shown rather good agreement with the theory for most substances measured. There were, however, substantial deviations from the theoretical magnitudes for measurements on low-resistivity Al films.[2,3] This Letter presents results of measurements on pure bulk type-I superconductors which disagree with the present prediction of the theory, and proposes a mechanism by which these results, those on similar systems, and the Al film results may be understood.

In magnetic measurements of the isotope effect in superconducting Zn[4] and Ga,[5] it was observed that some of the samples exhibited broad superconducting to normal (S-N) transitions in small magnetic fields. We have found that this paraconductivity is only observed in metallurgically and isotopically pure samples and in small magnetic fields (≤1 Oe). We observed measurable paraconductivity as much as 60 mK above the transition. Experimental details of the measurements are found in a previous paper.[4]

Figure 1 is a composite of S-N transitions of samples of Ga of relatively high metallurgical purity. The ordinates are the outputs of a mutual inductance detection system that was nulled when the samples were normal. Traces I, II, and III are transitions of Ga^{71} (99.61% Ga^{71}, 0.39% Ga^{69}) in 0.00, 0.52, and 1.04 Oe, respectively. Trace IV is the transition of natural Ga (60.0%

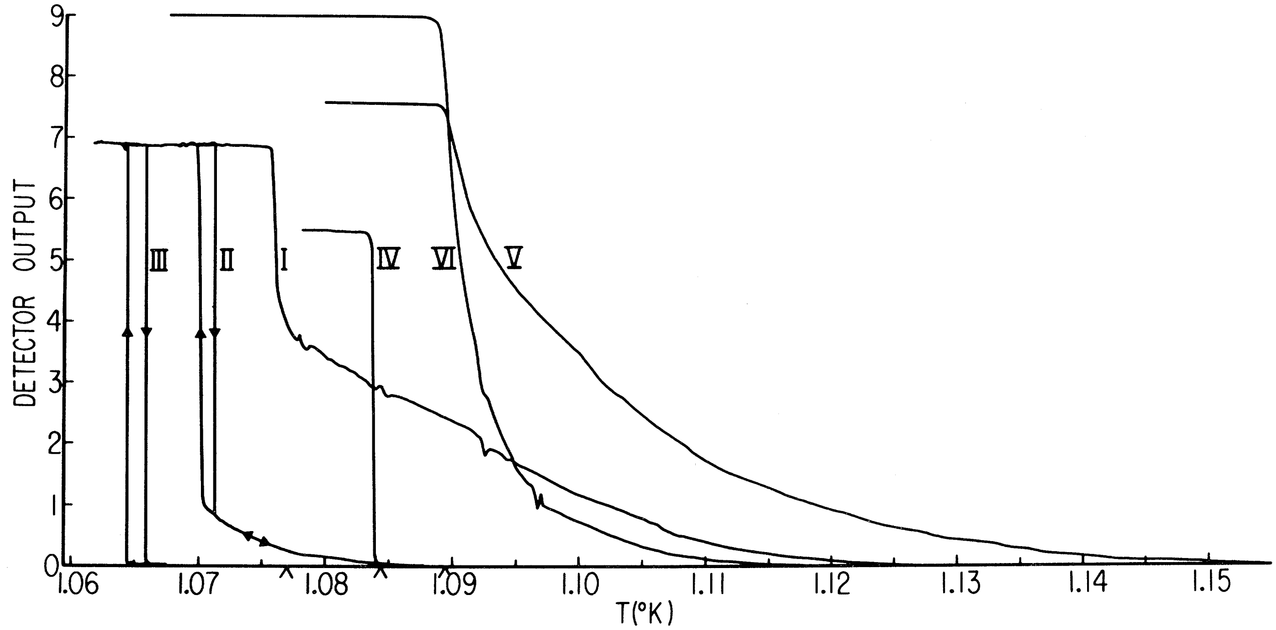

FIG. 1. Superconducting-to-normal transitions of samples of Ga in small magnetic fields. I, II, and III are transitions of 99.61% enriched Ga^{71} in 0.00, 0.52, and 1.04 Oe, respectively; IV is the transition of natural Ga in 0.00 Oe; and V and VI are the transitions of 99.77% enriched Ga^{69} in 0.00 Oe before and after the sample was damaged.

Ga^{69}, 40.0% Ga^{71}) in 0.00 Oe. The transitions of Ga^{69} (99.77% Ga^{69}, 0.23% Ga^{71}) were made in zero field before (V) and after (VI) the sample was damaged. Values of T_c (1.0770, 1.0845, and 1.0897) were determined by extrapolation of the critical-field curves to $H = 0.00$ for the three samples and are indicated by the carets at the baseline of Fig. 1.

The actual temperature dependence of the paraconductivity was not obtained, because the frequency of the measuring field (96 Hz, 0.01 Oe max) and the normal-state conductivity place the measurements in the anomalous skin-effect region, and the basic assumptions used in skin-effect calculations[6,7] are not valid for this system.

Traces I, II, and III indicate the strong dependence of the paraconductivity on the applied magnetic field. Trace II, which shows the coexistence of the paraconducting state and supercooling, indicates that the increased conductivity is not due to macroscopic superconducting regions. Traces V and VI demonstrate that the paraconductivity is greatly inhibited by crystalline imperfection. The magnitude of the influence of isotopic impurity on the paraconductivity is illustrated by the fact that our sample of Zn^{66} (98.8% Zn^{66}, 1.2% other Zn isotopes, and <1 at. ppm of other metallic impurities) showed almost no such effect. It should be noted that, except for Al, Nb (a type-II superconductor), and Bi (superconducting in amorphous films and high-pressure phases), all naturally occurring superconductors are isotopically mixed. Thus, behavior associated with isotopic purity should be found only in the above materials and in isotopically enriched samples.

Magnetic measurements by other workers of the S-N transitions in small fields have shown considerable broadening in pure single-crystal Al,[8] and in separated isotopes of Zn.[9] Detailed magnetic measurements on extremely pure natural Ga single crystals[10] reveal a transition that is 90% complete in an interval of $\sim 2\times 10^{-5}$ K. Recent measurements on bulk In[11] and Sn[12] have shown small enhanced diamagnetism above T_c in reasonable agreement with existing theory. The transitions of our isotopically mixed sample of natural Ga also showed this small effect.

Such clear corroboration of our results has not come from heat-capacity measurements which would indicate bulk rather than only surface effects. Zero-field transitions determined from heat capacities in Al[13,14] (~2 mK wide) are only wider than similar transitions in Sn[15] by a factor of 2 to 4.

We would point out that one significant difference between isotopically pure and mixed superconductors is the average phonon mean free path. Isotope scattering of phonons, a well-known phenomenon in the thermal conductivity of dielectric solids,[16] should also be important in metals. Such scattering of the phonons mediating superconductivity should limit pair lifetimes as does an applied magnetic field. Thompson[17] has suggested that in the absence of significant pair breaking, higher order diagrams than the first order ones considered by AL will become important. We would suggest that phonon mean free path effects may be important pair-lifetime limiting mechanisms.

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

[2] R. E. Glover, III, in Proceedings of the International Conference on the Science of Superconductivity, Stanford Univ., Stanford, Calif., August 1969 (to be published). This gives a review of experimental measurements of superconducting fluctuation effects.

[3] W. E. Masker and R. D. Parks, Phys. Rev. B (to be published).

[4] R. E. Fassnacht and J. R. Dillinger, Phys. Rev. 164, 565 (1967).

[5] R. E. Fassnacht and J. R. Dillinger, Phys. Letters 28A, 741 (1969).

[6] G. E. H. Reuter and E. H. Sondheimer, Proc. Roy. Soc. (London), Ser. A 195, 336 (1948).

[7] R. B. Dingle, Physica 19, 311 (1953).

[8] J. F. Cochran and D. E. Mapother, Phys. Rev. 111, 132 (1959); J. F. Cochran, D. E. Mapother, and R. Mould, Phys. Rev. 103, 1657 (1956).

[9] T. H. Geballe and B. T. Matthias, IBM J. Res. Develop. 6, 256 (1962).

[10] W. D. Gregory, Phys. Rev. 165, 556 (1968).

[11] J. P. Gollub, M. R. Beasley, R. S. Newbower, and M. Tinkham, Phys. Rev. Letters 22, 1288 (1969).

[12] R. Doll, Phys. Letters 29A, 534 (1969).

[13] D. C. Hopkins, private communication.

[14] D. C. Rohrer, H. Meyer, and R. C. Richardson, Z. Naturforsch. 18a, 130 (1963).

[15] J. F. Cochran, Ann. Phys. (N. Y.) 19, 186 (1962).

[16] E. g., P. D. Thacher, Phys. Rev. 156, 975 (1967).

[17] R. S. Thompson, in Proceedings of the International Conference on the Science of Superconductivity, Stanford Univ., Stanford, Calif., August 1969 (to be published); R. S. Thompson, Phys. Rev. B 1, 327 (1970).

NONLOCAL CHARACTERISTICS OF THE BULK UPPER CRITICAL FIELD OF NIOBIUM

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Measurements of the anisotropic critical field H_{c2} of Nb at temperatures down to 0.06 K show that its temperature dependence contains a logarithmic factor as predicted by Gor'kov on the basis of a nonlocal theory for an isotropic type-II superconductor. The critical field extrapolated to zero temperature, when averaged over all crystallographic directions and normalized to the slope of the critical field for $T \approx T_c$, has an enhancement close to that predicted by the Hohenberg-Werthamer calculation for the nonlocal effects of Fermi-surface anisotropy, as evaluated by Mattheiss for a band model of niobium.

Considerable interest has been focused upon the temperature dependence of the upper critical field H_{c2} of pure Nb, since this metal is an elemental example of a pure type-II superconductor, yet the bulk critical field is known[1] to deviate substantially from the rigorous solution obtained by Helfand and Werthamer[2] for the linearized Ginzburg-Landau-Abrikosov-Gor'kov (GLAG) theory. Observations by Tilley, van Gurp, and Berghout and by Reed et al.[4] showed that H_{c2} is anisotropic and, therefore, must reflect "real metal" effects. Reed et al.[4] found that the temperature dependence of the anisotropy near the critical temperature T_c was qualitatively in agreement with the calculation of Hohenberg and Werthamer[5] (hereafter, HW), which attributed such effects to Fermi-surface anisotropy as included in a first-order nonlocal correction to the GLAG theory. However, no quantitative test of the theory was possible, nor was the temperature dependence of H_{c2} at low temperatures predicted. More recently it has been suggested by Sung and Wong[6] that two-band effects, such as are indicated at low temperatures by specific-heat[7] and thermal-conductivity[8] measurements, might influence $H_{c2}(T)$ if interband coupling parameters are properly chosen; however, again, no quantitative test of this theory has been made.

We report in this Letter that H_{c2} values for the applied field parallel to the [001], [110], and [111] directions and for temperatures as low as