

## Concerning the Ratio $H_{c3}/H_{c2}$ in Superconducting Nb and Nb-O Solutions\*

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The Saint-James-de Gennes relationship between  $H_{c2}$  and  $H_{c3}$  in type II superconductors has been verified in Nb and Nb-O interstitial solid solutions. The observed ratio  $H_{c3}/H_{c2}=1.7_0$  is found to be independent of  $\kappa$ , the Ginsburg-Landau parameter, for niobium-containing oxygen in concentrations below the solubility limit at the temperature of introduction. Critical-current data  $I_c$  versus  $H$  above  $H_{c2}$ , analyzed in terms of a dimension of the superconducting sheath, (e.g.,  $J_s=I_c$  per cm width), reveal sharp breaks at  $H_{c3}$  when the material becomes normal. For a cold-worked specimen, two portions of the  $J_s(H)$  curve may be identified, meeting at a definite field corresponding to  $H_{c3}$  calculated from theory. The upper portion of the curve first meets the normal state curve at a field  $H_n$ . The ratio  $H_n/H_{c2}$  is larger for cold-worked niobium than for cold-worked niobium-oxygen alloys.

### INTRODUCTION

RECENTLY, Saint-James and de Gennes<sup>1</sup> have shown that in type II superconductors in parallel fields, nucleation of a superconducting sheath will occur at the field  $H_{c3}=1.695H_{c2}$ , where  $H_{c2}=\kappa\sqrt{2}H_c$ .<sup>2,3</sup>  $H_c$  is the thermodynamic critical field and  $\kappa$ , the Ginsburg-Landau parameter. For the surface perpendicular to the field, the sheath of superconductivity should not exist beyond  $H_{c2}$ , Abrikosov's upper critical field. These predictions have been well substantiated by experiment, primarily in superconducting, substitutional solid solutions.<sup>4-8</sup>

In interstitial solid solutions where sample homogeneity is more difficult to achieve, it is possible for the specimen surface to be characterized by one  $\kappa$  and the interior by another, artificially forcing the apparent ratio  $H_{c3}/H_{c2}$  in either direction.<sup>9</sup> The interstitial solutions are essentially supersaturated and because the interstitial atom has great mobility, it can easily segregate to structural irregularities such as dislocations.

In this report we show that the ratio  $H_{c3}/H_{c2}$  observed in pure niobium and niobium containing oxygen in a concentration below the solubility limit at the temperature of introduction is in good agreement with the theoretical value proposed by Saint-James and de Gennes and found to be independent of concentration and/or  $\kappa$ . The influence of cold work, quenching, and

strain-aging on the ratio and on  $H_n$ , a field larger than  $H_{c3}$ , is also cited.

### EXPERIMENTAL

The experimental details have been described elsewhere.<sup>9</sup> The specimens were rectangular in shape, made by pressing niobium<sup>10</sup> between highly polished stainless steel blocks. The ribbons were outgassed and annealed at a temperature 1800–2000°C for 24 h in vacuum less than  $1\times 10^{-7}$  mm Hg. They were  $7.1_1\times 10^{-2}$  cm wide by  $7.6_2\times 10^{-3}$  cm thick. A ribbon specimen of  $\text{Nb}_{0.9916}\text{O}_{0.0084}$  was prepared by adding oxygen at about 1100°C to a previously outgassed niobium ribbon; then homogenized at 1200°C for several hours. This sample was  $1.1_4\times 10^{-1}$  cm wide and  $1.9_1\times 10^{-2}$  cm thick.

Two field orientations were examined: (1) field parallel to the wide side of the sample ( $H\parallel\text{w.s.}$ ), and (2) field perpendicular to the wide side ( $H\perp\text{w.s.}$ ). In either case,  $H$  was perpendicular to the direction of current flow. The two field orientations were made precisely reproducible by mounting the ribbon in a form permanently fixed between the pole pieces of a small  $\text{Nb}_2\text{Zr}$  wire-wound, iron-core magnet.

### RESULTS AND DISCUSSION

Measurements of longitudinal voltages in the direction of  $J$  as a function of applied field for a given current and for field orientation (a)  $H\parallel\text{w.s.}$  and (b)  $H\perp\text{w.s.}$  are illustrated in Fig. 1 for  $\text{Nb}_{0.9916}\text{O}_{0.0084}$ . In both cases, the sample has two opposite surfaces parallel to the field, although the demagnetization coefficient is less favorable when  $H\perp\text{w.s.}$  and the total surface parallel to the field is smaller. For the two field directions studied,  $H_{c3}$  is evaluated when the potential first attains the normal-state value. This method of determining  $H_{c3}$  has some uncertainty since the normal voltage is approached asymptotically.

An alternative measurement is the determination of critical current  $I_c$  as a function of field, choosing the critical current as that current which gives an arbitrarily

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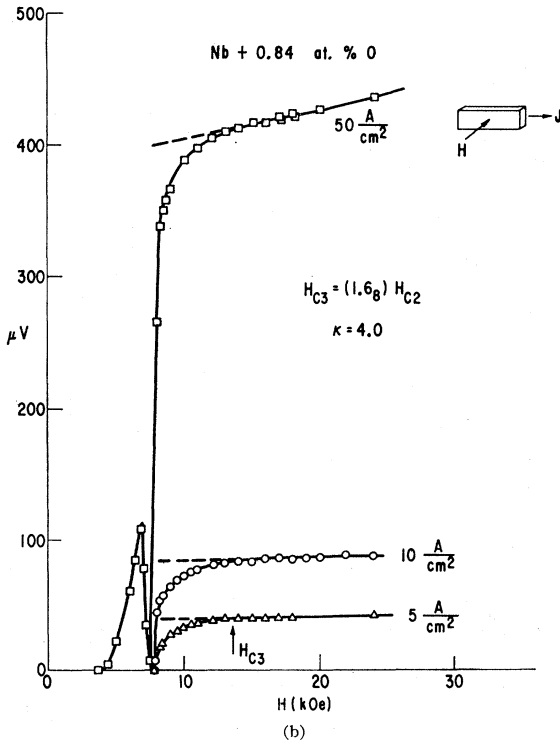
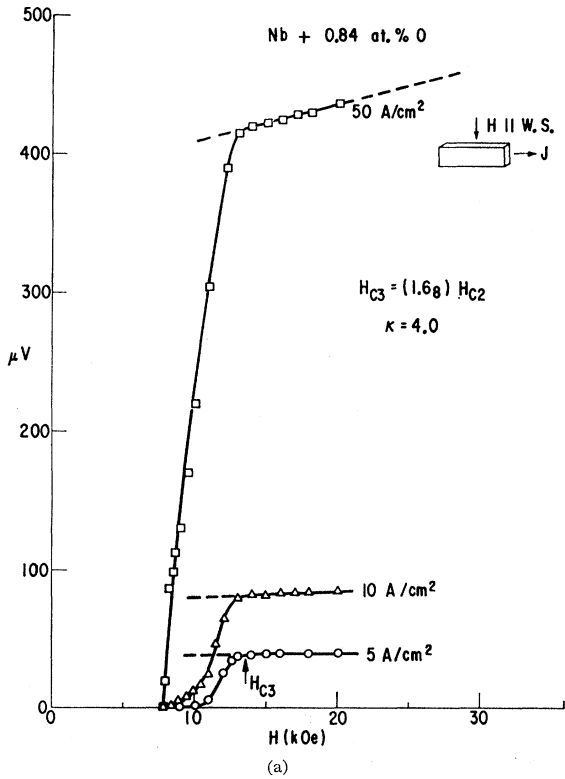


FIG. 1. The longitudinal voltage, along direction of  $J$ , observed as a function of current for a rectangular specimen  $Nb_{0.9916}O_{0.0084}$  ( $1.14 \times 10^{-1}$  cm wide  $\times 1.91 \times 10^{-2}$  cm thick). Distance between potential leads,  $7 \times 10^{-1}$  cm. (a) Field parallel to the wide side ( $H || w.s.$ ). (b) Field perpendicular to the wide side ( $H \perp w.s.$ ).  $H_{c3} \approx 13.0$  kOe,  $H_{c2} = 7.74$  kOe (magnetization),  $T = 4.2^\circ K$ .

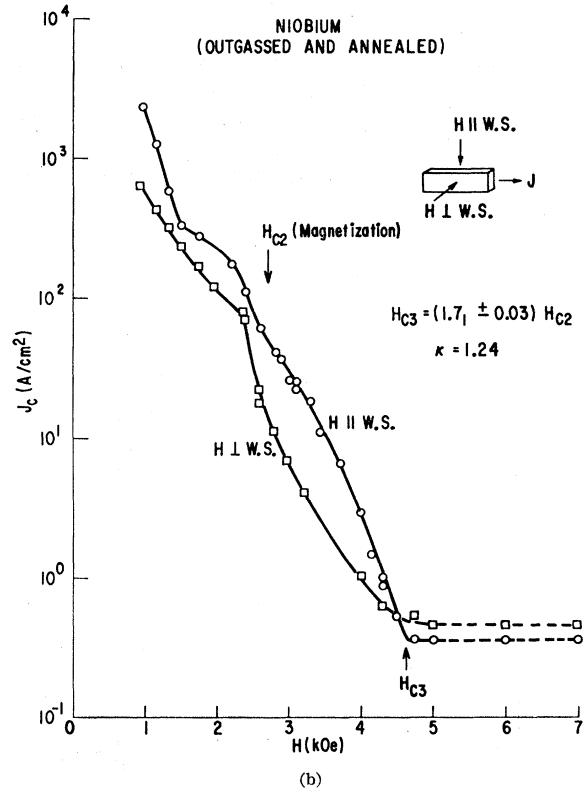
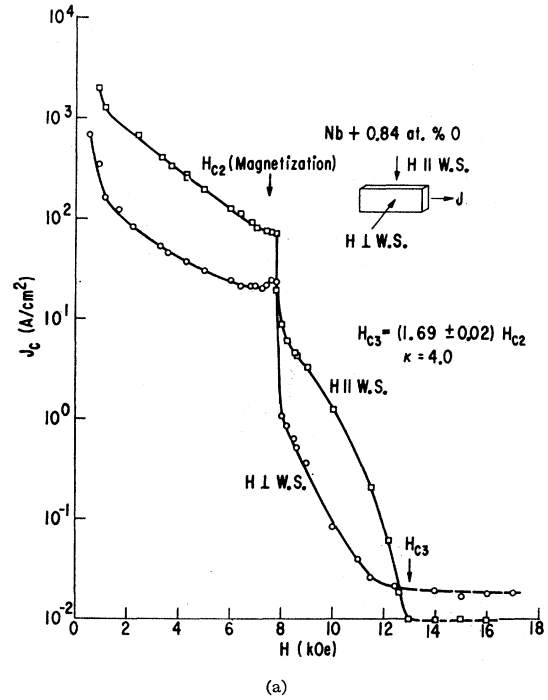


FIG. 2. The critical current  $J_c$  based on the cross-sectional area of the sample. (a) Specimen  $Nb_{0.9916}O_{0.0084}$  ( $1.14 \times 10^{-1}$  cm wide  $\times 1.91 \times 10^{-2}$  cm thick). (b) Nb ( $7.1 \times 10^{-2}$  cm wide  $\times 7.62 \times 10^{-3}$  cm thick).  $H_{c3} = 4.62$  kOe,  $H_{c2} = 2.70$  kOe (magnetization),  $T = 4.2^\circ K$ . For fields above  $H_{c3}$  and for  $H || w.s.$  the broken horizontal straight line represents normal state behavior.

chosen voltage. An arbitrarily chosen voltage is the minimum detectable voltage above noise level. For  $H \parallel$  w.s., this voltage decision level was found to be minimum, equal to approximately  $0.01 \mu\text{V}$ . For cold-worked ribbons, the minimum voltage above noise level was consistently higher, attaining maximum values of  $0.1 \mu\text{V}$  when  $H \perp$  w.s. These variations in noise level, although not understood, are too small to alter the conclusions following below. Some typical  $I_c$  data in terms of critical current density,  $J_c$  (defined as the critical current per cross sectional area of the sample) as a function of field, are summarized in Fig. 2(a) and 2(b) for  $\text{Nb}_{0.9916}\text{O}_{0.0084}$  and pure Nb, respectively. Above  $H_{c2}$ ,  $J_c$  is seen to fall with increasing  $H$  until a value independent of field is reached. For  $H \parallel$  w.s. this value of current density corresponds to that current in the normal state giving the arbitrary decision level. In this region, the sample is not superconducting. For  $H \perp$  w.s., the  $J_c$  versus  $H$  curve approaches the normal state curve rather decisively permitting a more accurate evaluation of  $H_{c3}$ . The ratio,  $H_{c3}/H_{c2}$ , evaluated from these data, is independent of  $\kappa$  in these two samples and in good agreement with the Saint-James and de Gennes theoretical value.  $H_{c2}$  has been determined from magnetization data.

If a superconducting sheath exists in the region  $H_{c2} \leq H \leq H_{c3}$ , as predicted, it would be more appropriate to analyze  $I_c$  data in terms of some dimension of this sheath pertaining only to the surfaces parallel to the applied field for a given orientation. A simple analysis would be the evaluation of the critical current flow associated per unit width of sheath, or  $J_s = I_c/\text{cm}$

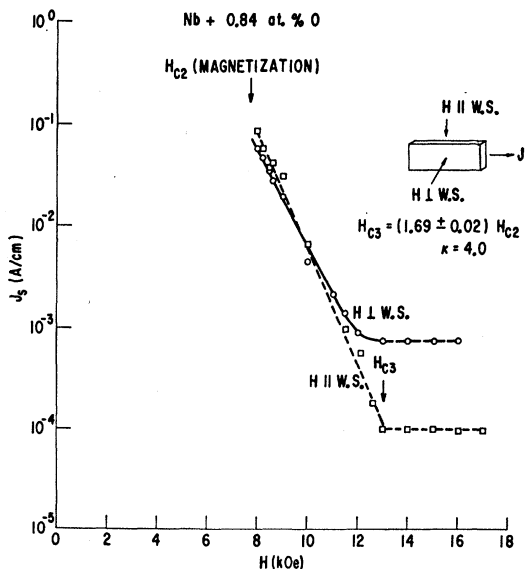


FIG. 3. The critical current  $J_s$  calculated from  $I_c$  per unit width of superconducting sheath. Thickness of sheath assumed independent of field and direction for  $\text{Nb}_{0.9916}\text{O}_{0.0084}$  ( $1.41 \times 10^{-1}$  cm wide  $\times 0.91 \times 10^{-2}$  cm thick).  $T = 4.2^\circ\text{K}$ . Horizontal curve represents normal-state behavior.

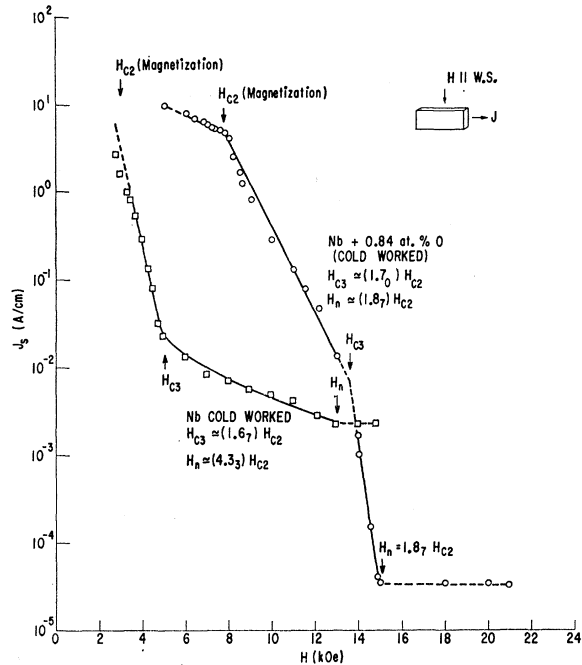


FIG. 4. The critical current  $J_s$  calculated from  $I_c$  per unit width of superconducting sheath for (a) cold-worked Nb ( $1.37 \times 10^{-1}$  cm wide  $\times 8.8_3 \times 10^{-3}$  cm thick), (b) cold-worked  $\text{Nb}_{0.9916}\text{O}_{0.0084}$  ribbons ( $1.1_9 \times 10^{-1}$  cm  $\times 1.5_2 \times 10^{-2}$  cm);  $T = 4.2^\circ\text{K}$ .

width.  $J_s$  evaluated in this manner is summarized in Fig. 3 as a function of  $H$  for  $\text{Nb}_{0.9916}\text{O}_{0.0084}$  and for  $H \parallel$  w.s. and  $H \perp$  w.s. For  $H \parallel$  w.s., the curve above  $H_{c2}$ , approximately linear on the semilog plot, forms an abrupt break with the horizontal normal-state curve. The field at which this break occurs corresponds to  $H_{c3}$ , calculated. Within experimental error, the approximate agreement in  $J_s(H)$  for the two field orientations between  $H_{c2}$  and  $H_{c3}$  is evident. Similar results have also been obtained for pure niobium. This behavior indicates that the surfaces perpendicular to the applied field are, apparently, not contributing any remanent superconductivity above  $H_{c2}$  in agreement with theory.<sup>1</sup>

The results summarized in Fig. 4 were obtained on Nb and  $\text{Nb}_{0.9916}\text{O}_{0.0084}$  ribbons cold worked by compression and again evaluating  $J_s$  for  $H \parallel$  w.s. Above  $H_{c2}$  and fields below the normal-state behavior, two distinct portions of the curve may now be identified. The field at the junction of these two segments corresponds to  $H_{c3}$  calculated from the Saint-James and de Gennes theory. The slope of the linear curve,  $dJ_s/dH$ , for either sample is approximately the same as that obtained from the data prior to the deformation. It is larger when  $\kappa$  is smaller. The absolute value of  $J_s$ , however, appears to be larger after being cold worked.

The upper portion of the curve above  $H_{c3}$  terminates at the normal state at a field  $H_n$ .  $H_n$  is larger relative to  $H_{c2}$  for the cold-worked niobium than for cold-worked  $\text{Nb}_{0.9916}\text{O}_{0.0084}$  solid solution.

Deformation produces inhomogeneous distribution of dislocations; therefore, an inhomogeneous mean free path and an inhomogeneous  $\kappa$ . This can cause  $H_n/H_{c2}$  ratios greater than the ideal value 1.69. However, in an alloy, the mean free path is controlled primarily by solute concentration and variations in dislocation density cause much smaller variations in  $\kappa$  than in pure material. Therefore, in the Nb-O solution one would expect the ratio  $H_n/H_{c3}$  to be smaller than that observed in Nb.<sup>11</sup>

In an experiment on varying the degree of segregation at dislocations by (a) quenching, (b) cold working, and (c) strain aging (heating 3 h, 170°C), marked changes

<sup>11</sup> J. D. Livingston (private communication).

have been observed in the resistivity in the mixed state between  $H_{c1}$  and  $H_{c2}$  (see Figs. 16 and 17, Ref. 9) for Nb<sub>0.993</sub>O<sub>0.007</sub>. After quenching,  $H_{c3}/H_{c2}=1.71$ . Cold working the quenched sample increased  $H_n$  to approximately 1.8<sub>3</sub>  $H_{c2}$ , while strain aging may increase  $H_n$  slightly ( $H_n \approx 1.8_8 H_{c2}$ ).

Analysis of  $I_c$  data above  $H_{c2}$  in terms of  $J_s$  may help to elucidate some of the higher values reported for  $H_{c3}$  in type II superconductors (e.g., see Ref. 7).

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## Perturbation Approach to the Diffraction of Electromagnetic Waves by Arbitrarily Shaped Dielectric Obstacles\*

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A perturbation method is developed to consider the problem of the diffraction of electromagnetic waves by an arbitrarily shaped dielectric obstacle whose boundary may be expressed in the general form, in spherical coordinates,  $r_p = r_0[1 + \delta f_1(\theta, \phi) + \delta^2 f_2(\theta, \phi) + \dots]$  where  $r_0$  is the radius of an unperturbed sphere and  $f_n(\theta, \phi)$  are arbitrary, single-valued and analytic functions.  $\delta$  is chosen such that

$$\sum_{n=1}^{\infty} |\delta^n f_n(\theta, \phi)| < 1, \quad 0 \leq \theta \leq \pi, \quad 0 \leq \phi \leq 2\pi.$$

Detailed analysis is carried out to the first order in  $\delta$ . Procedures to obtain higher order terms are also indicated. The perturbation solutions are valid for the near zone region of the obstacle as well as for the far zone region and they are applicable for all frequencies. Possible applications of this perturbation technique to elementary-particle scattering problems and other electromagnetic scattering problems are noted.

### I. INTRODUCTION

THE exact solution of the problem of the diffraction of electromagnetic waves by an obstacle of given shape and electromagnetic properties can be found only in a few cases.<sup>1,2</sup> For example, the diffraction of waves by a conducting or dielectric sphere, by dielectric coated spheres and by a perfectly conducting disk are the few three-dimensional problems that have been solved rigorously. The need for approximate methods to treat the more general cases of diffraction from arbitrarily shaped obstacles is quite apparent. The variational principles<sup>3,4</sup> provide a very powerful tool in obtaining

an approximate expression for the scattering cross section; but it is not possible to derive from the variational principles a description of the electromagnetic fields. Furthermore, the success of the variational approach depends to a great extent on the trial function. At low frequencies, the Rayleigh method is very useful.<sup>5,6</sup> However, the solutions of Laplace's equation are still required. At very high frequencies, the treatment of diffraction problems by geometric and physical optics techniques developed by Fock<sup>7</sup> and Keller<sup>8</sup> is very successful. An approximate or perturbation method in the medium frequency range still remains to be found.

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