## Comments and Addenda

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## Variation of the Ratio $H_{c3}/H_{c2}$ in the Immediate Vicinity of $T_c^+$

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We have measured the ratio  $H_{c3}/H_{c2}$  for tin-rich Sn-In alloys and for indium-rich In-Bi alloys in the immediate vicinity of  $T_e$ . For the Sn-In system this ratio decreases as  $T_e$  is approached. Similar behavior has been recently reported by Ostenson and Finnemore for pure Nb. The In-Bi alloys, however, show a marked increase in  $H_{c3}/H_{c2}$  as the critical temperature is approached.

 ${f R}$  ECENTLY, Ostenson and Finnemore<sup>1</sup> have reported a substantial decrease in the ratio of the surface critical field  $H_{c3}$  to the bulk critical field of the mixed state  $H_{c2}$  for pure Nb in the immediate vicinity of  $T_c$ . We report in this paper measurements of  $H_{c3}$ and  $H_{c2}$  very close to  $T_c$  for type-II superconductors of the Sn-rich Sn-In system and the In-rich In-Bi system. A decrease in  $H_{c3}/H_{c2}$ , similar to that reported for Nb, is observed in the Sn-In alloys as  $T_c$  is approached. The measured alloys of the In-Bi system, however, show



FIG. 1. Typical magnetic transition for  $\text{Sn}_{0.95}$  In<sub>0.05</sub> alloy at a reduced temperature t=0.943. Upper and lower curves correspond to  $H_{c3}$  and  $H_{c2}$ , respectively. The upper curve has been measured for **H** parallel to the plate and the lower curve for **H** perpendicular.

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188 (1969).

an increase in the ratio  $H_{c3}/H_{c2}$ , which becomes higher  $(\sim 2.5)$  than the theoretical value (1.695).<sup>2</sup>

We have measured Sn-In alloys with 5- and 6-at.%In, and In-Bi alloys with 2.9- and 2-at.% Bi. The samples, thin plates 0.1 mm thick, 4 mm wide, and 10 mm long, were prepared by rolling the alloy between stainless-steel rollers. The alloys were prepared by melting and vibrating the components for several hours. The rolled samples were annealed for 24 h before performing the measurements. The critical fields were determined by measuring the mutual inductance of a pair of coils with the sample between them. The ac magnetic field produced by the primary of those coils was kept smaller than 0.1 Oe. The frequency used was 1000 Hz. The surface critical field  $H_{c3}$  was determined with an external dc magnetic field H parallel to the large surface of the sample, while  $H_{c2}$  was assumed to be the critical field for **H** perpendicular to that surface. We believe that the surface superconductivity at the sample edges is too small to affect our measurements. This belief is supported by the fact that for  $T \ll T_c$  values of  $H_{c3}/H_{c2}$  quite close to the theoretical one are obtained by this method. In the course of the measurements, the amplitude of the ac probing field was changed by a factor of 2 and its frequency reduced to 200 Hz. No dependence of  $H_{c3}$  and  $H_{c2}$  on these parameters was found. The change in mutual inductance was measured with a conventional bridge<sup>3</sup> and plotted on an x-yrecorder as a function of dc magnetic field. The field Hwas swept at a constant rate chosen slow enough to give retraceable curves for increasing and decreasing fields.

<sup>&</sup>lt;sup>2</sup> D. Saint James and P.-G. de Gennes, Phys. Letters 7, 306

<sup>(1963).</sup> <sup>\*</sup> W. L. Pillinger, P. S. Jastram, and J. G. Daunt, Rev. Sci. Instr. 29, 159 (1958).



FIG. 2. Values of  $H_{c3}$ ,  $H_{c2}$ , and the ratio  $H_{c3}/H_{c2}$  for Sn<sub>0.95</sub> In<sub>0.05</sub> alloy. The critical temperature chosen for this alloy was  $T_c=3.625$ .

Possible errors due to uncertainties in the position of the sample<sup>4</sup> were minimized by measuring the nominal ratio  $H_{c3}/H_{c2}$  for various azimuthal orientations of H. The angle for which a maximum in  $H_{c3}/H_{c2}$  occurs (**H** parallel to the sample surface) could be determined to better than 1°. In a similar manner, we also checked the absence of errors due to the earth's magnetic field.

We show in Fig. 1 the magnetic transition observed for a  $Sn_{0.95}$  In<sub>0.05</sub> alloy at a reduced temperature t=0.943. The shape of this transition is typical of all Sn-In samples measured; the transition of the In-Bi alloys is somewhat wider. The three most obvious criteria for the definition of the critical field correspond to the points labeled A, B, and C in Fig. 1, i.e., the middle point in the vertical axis (A), the linear extrapolation to join the mutual inductance of the normal state (B), and the rather poorly defined tail point (C) at which all traces of superconductivity seem to have disappeared. We have analyzed our data with these three criteria and found that, to a constant scaling factor, the functional dependence of  $H_{c2}$  and  $H_{c3}$  on temperature is independent of the criterion used. Therefore, the ratio  $H_{c3}/H_{c2}$  does not depend on this criterion, provided the same criterion is used for both  $H_{c3}$  and  $H_{c2}$ . If different criteria are used for  $H_{c3}$  and  $H_{c2}$ , the ratio  $H_{c3}/H_{c2}$  is changed by a constant factor of the order of 1. The use of the same criterion is necessary in order to obtain a value of  $H_{c3}/H_{c2}$  close to the theoretical ratio<sup>2</sup> for  $T \ll T_c$ . For the data presented here, we have chosen the points B as the critical fields.

We show in Fig. 2 the values of  $H_{c3}$  and  $H_{c2}$  obtained for Sn<sub>0.95</sub> In<sub>0.05</sub> near  $T_c$ . The insert in this figure shows



FIG. 3. Values of  $H_{c3}$ ,  $H_{c2}$ , and the ratio  $H_{c3}/H_{c2}$  for In<sub>0.98</sub> Bi<sub>0.02</sub>. The critical temperature chosen was  $T_c=3.845$ .

the temperature dependence of the ratio  $H_{c3}/H_{c2}$  for this alloy, which is rather similar to the corresponding ratio observed by Ostenson and Finnemore<sup>1</sup> for Nb. The decrease in  $H_{c3}/H_{c2}$  starts at the reduced temperature t=0.92, while  $H_{c3}$  and  $H_{c2}$  can be fitted to straight lines up to t=0.98. The decrease in  $H_{c3}/H_{c2}$  is associated with the fact that the straight lines do not intercept the horizontal axis at the same point.

Figure 3 shows  $H_{c3}$ ,  $H_{c2}$ , and  $H_{c3}/H_{c2}$  for an In-Bi alloy (2-at.%) Bi). The ratio  $H_{c3}/H_{c2}$  increases for t > 0.91. For  $0.91 \le t \le 0.97$  this increase is due to the fact that the linear variations of  $H_{c3}$  and  $H_{c2}$  do not extrapolate to the same temperature on the horizontal axis. The observation is similar to that mentioned above for the Sn-In alloy, but in Fig. 3 an increase in  $H_{c3}/H_{c2}$  results as  $T_c$  is approached due to the fact that  $H_{c3}$  extrapolates linearly to a temperature higher than  $H_{c2}$ . For t > 0.97,  $H_{c3}/H_{c2}$  keeps increasing, but  $H_{c3}$  and  $H_{c2}$  do not vary linearly with T. The temperature dependence of  $H_{c3}/H_{c2}$  shown in Figs. 2 and 3 seems to be characteristic of the corresponding alloy systems. Samples of Sn0.94 In0.06 and In0.971 Bi0.029 gave essentially the same type of dependence of  $H_{c3}/H_{c2}$ on temperature as that shown in Figs. 2 and 3, respectively.

A theoretical interpretation of the temperature variation of  $H_{c3}/H_{c2}$  near  $T_c$  reported here is not available. Further experiments to clarify the concentration dependence of this variation for these and other alloy systems are in progress.

Note added in proof: Recently, we have measured  $H_{c3}/H_{c2}$  for Sn-In alloys as a function of sample thickness. We observed that the anomalies in  $H_{c3}/H_{c2}$  discussed here disappear for thickness  $\simeq 0.02$  mm. The detailed results will be published elsewhere.

<sup>&</sup>lt;sup>4</sup> The surface critical field is known to decrease drastically if H is not exactly parallel to the surface. See, for instance, P. Burger, G. Deutscher, E. Guyon, and A. Martinet, Phys. Rev. 137, A853 (1965).