Brief Reports

Brief Reports are short papers which report on completed research which, while meeting the usual **Physical Review** *standards of scientific quality, does not warrant a regular article. (Addenda to papers previously published in the* **Physical Review** *by the same authors are included in Brief Reports.) A Brief Report may be no longer than 3½ printed pages and must be accompanied by an abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Superconductivity and magnetism of bcc Cr-Ru alloys

Y. Nishihara, Y. Yamaguchi, and M. Tokumoto
Electrotechnical Laboratory, Sakura-mura, Ibaraki 305, Japan

K. Takeda
Faculty of Engineering, Kyushu University, Fukuoka 812, Japan

K. Fukamichi
The Research Institute for Iron, Steel and Other Metals, Tohoku University, Sendai 980, Japan
(Received 31 March 1986)

The superconducting and magnetic properties of bcc Cr-Ru alloys with between 14 and 25 at.% Ru were studied via measurements of electrical resistivity, specific heat, and magnetic susceptibility. The temperature dependence of the specific heat revealed that bcc Cr-Ru alloys with a concentration higher than ~17 at.% Ru exhibit superconductivity below ~2 K. The magnetic susceptibility of these alloys shows a peak around 170 K. The present result suggests that superconductivity coexists with itinerant-electron antiferromagnetism in bcc Cr-Ru alloys.

I. INTRODUCTION

The Cr-Ru system has a bcc structure up to ~30 at.% Ru. The superconducting properties of Cr-Ru alloys were investigated by Matthias *et al.*[1] They concluded that the solid solution of Ru in bcc Cr is not superconducting above 0.3 K and that annealed Cr_3Ru with the $A15$ structure has a superconducting transition temperature of 3.3 K. The magnetic transition temperature of bcc Cr-Ru alloys was measured by Booth.[2] The antiferromagnetic transition temperature increases from ~300 to ~600 K with increasing Ru concentration up to ~2 at.% Ru. Above ~6 at.% Ru the magnetic transition temperature decreases with increasing Ru concentration. There was no report on the concentration region higher than 15 at.% Ru.

The antiferromagnetic transition temperature of the Cr-Re system was studied by many workers.[2-4] The Cr-Re system shows behavior similar to that of the Cr-Ru system in the magnetic transition temperature and furthermore the former exhibits superconductivity and antiferromagnetism in the region higher than 18 at.% Re.[2-4] In order to search for the difference between the Cr-Re and Cr-Ru systems, we have studied the electrical and magnetic properties of bcc Cr-Ru alloys and found that the Cr-Ru system also exhibits superconductivity below ~2 K in the region higher than ~17 at.% Ru. This paper gives experimental results on the superconducting and magnetic properties of bcc Cr-Ru alloys.

II. EXPERIMENTAL RESULTS

$Cr_{100-x}Ru_x$ $(14 \leqq x \leqq 25)$ alloys were prepared by arc melting, using 99.99%-pure Cr and 99.9%-pure Ru. X-ray diffraction of the as-cast alloys gave the bcc pattern without extra phases. The lattice constant of the bcc alloys increases with increasing Ru concentration as shown in Fig. 1. The $A15$ structure of Cr_3Ru was obtained by annealing the alloy at 800 °C for two weeks. This sample

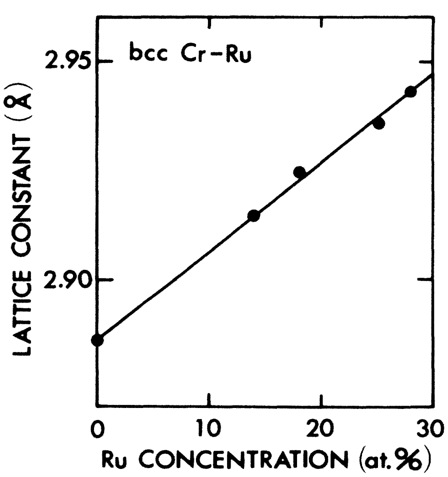

FIG. 1. Lattice constant of bcc Cr-Ru alloys.

is superconducting below 3.25 K and has a lattice constant of 4.677 ± 0.001 Å.

Figure 2 shows the temperature dependence of the upper critical field H_{c2} determined from the midpoint of the resistivity change. Matthias *et al.* reported[1] that the solid solution of Ru in bcc Cr is not superconducting above 0.3 K, since the predominant phase of the superconducting Cr-Ru alloy has the *A*15 structure. However, the present result demonstrates that superconductivity is observed even in the Cr-Ru alloys of pure bcc phase. To investigate whether the superconductivity comes from the bulk of the sample or not, we have measured the temperature dependence of the specific heat of the bcc $Cr_{75}Ru_{25}$ alloy which corresponds to the same Ru concentration as the *A*15 compound of Cr_3Ru.

Figure 3 shows the temperature dependence of the specific heat capacity C of bcc $Cr_{75}Ru_{25}$ in zero external field. The value of C increases exponentially with increasing temperature from 0.8 K and exhibits a peak at $T_c = 1.87$ K. From the plot of C/RT vs T^2, where R is the gas constant, we find that the specific heat jump at T_c is $\Delta C/\gamma T_c \sim 1.28$, which is $\sim 90\%$ of the value estimated from the BCS theory. Since this sample is in the pure bcc phase, the result indicates that the bcc $Cr_{75}Ru_{25}$ exhibits BCS-type superconductivity below T_c. The transition width at T_c is less than 0.05 K, which is much smaller than the width of ~ 0.5 K of Cr-Re alloys.[4,5] The electrical resistivity of this sample becomes zero below ~ 2.4 K $(= T_c^R)$, as shown in Fig. 2. However, there is no anomaly in the specific heat between 2.4 and 1.9 K within 2% of the peak value.

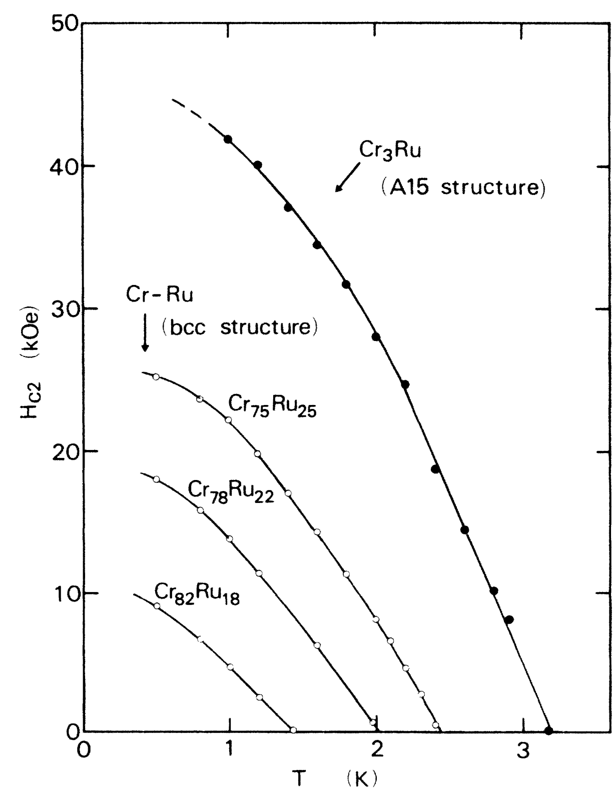

FIG. 2. Temperature dependence of the upper critical field of bcc Cr-Ru alloys and Cr_3Ru with the *A*15 structure.

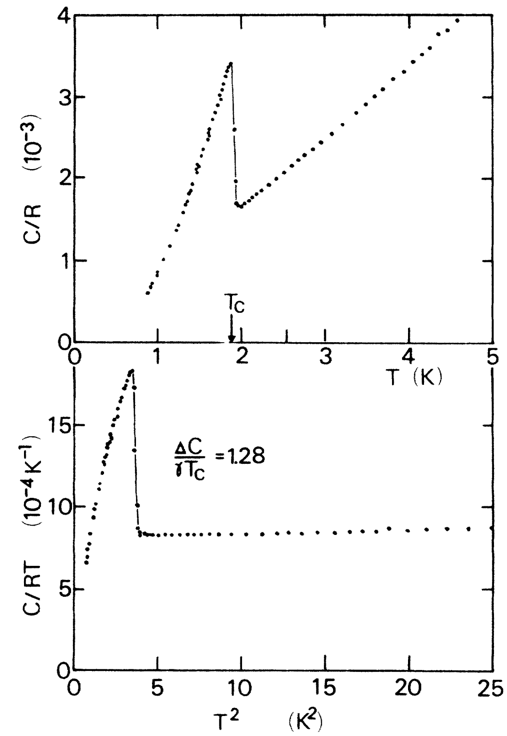

FIG. 3. Temperature dependence of the specific heat of bcc $Cr_{75}Ru_{25}$.

The magnetization curves of bcc $Cr_{75}Ru_{25}$ were measured below 2.4 K. The initial magnetization exhibits perfect diamagnetism as shown in the inset of Fig. 4. The lower critical field H_{c1} estimated from these curves shows the two-step increase with decreasing temperature. The value of H_{c1} is less than 10 Oe between T_c^R and T_c and in-

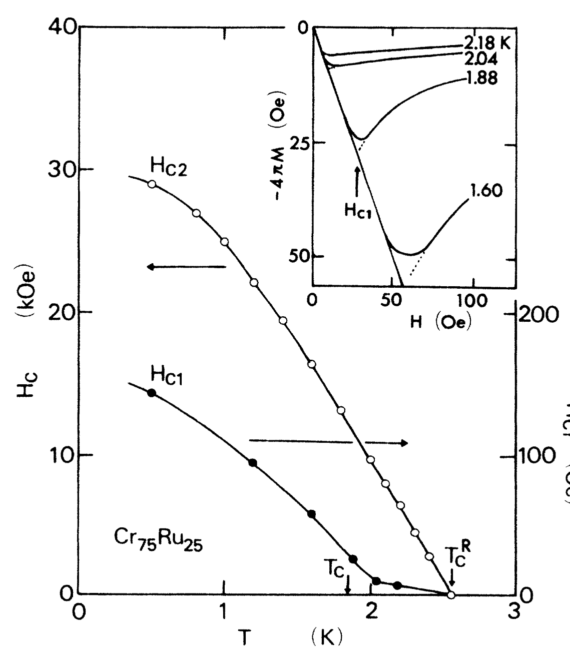

FIG. 4. Lower and the upper critical fields of H_{c1} and H_{c2} of bcc $Cr_{75}Ru_{25}$. The inset shows the magnetization curves below 100 Oe.

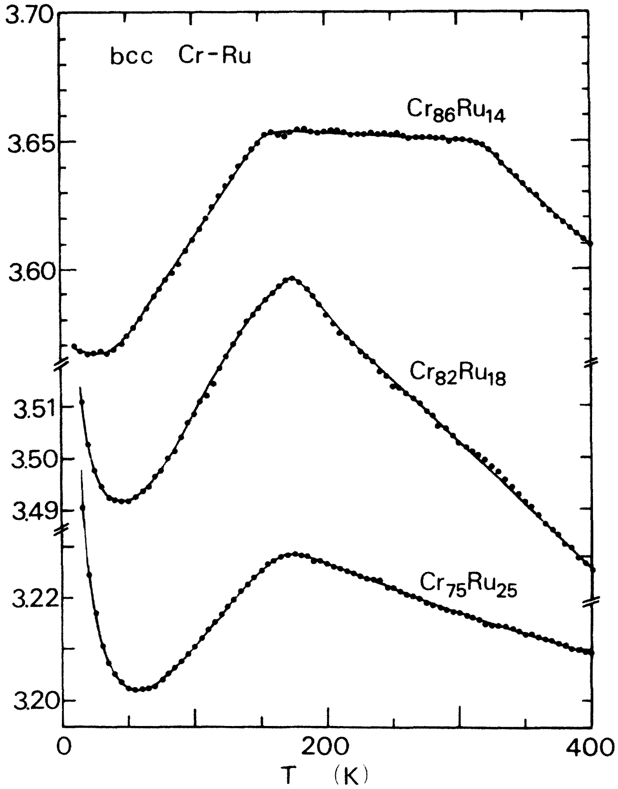

FIG. 5. Temperature dependence of the magnetic susceptibility of bcc Cr-Ru alloys.

creases rapidly below T_c up to $\sim$150 Oe. However, the upper critical field obtained from the resistivity measurement shows no anomaly at T_c.

Figure 5 shows the temperature dependence of the magnetic susceptibility of bcc Cr-Ru alloys. The susceptibility decreases with increasing Ru concentration and that of $Cr_{86}Ru_{14}$ shows two kinks at around 160 and 320 K. With increasing Ru concentration only the low-temperature peak remains. The position of the peak is almost independent of the Ru concentration within $\pm$10 K.

III. DISCUSSION

The superconducting and magnetic transition temperatures are illustrated in Fig. 6. The antiferromagnetic transition temperatures for the alloys with concentrations lower than 14 at.% Ru are those from Refs. 2 and 6. The susceptibility of bcc $Cr_{86}Ru_{14}$ shows two kinks in the vicinity of 160 and 320 K. The high-temperature kink continuously connects with the antiferromagnetic transition temperature obtained by Booth[2] in the lower-Ru-concentration region. In the alloy with 18 at.% Ru the susceptibility peak is observed only at around 170 K and the superconducting state appears at low temperatures. The peak in susceptibility shows that antiferromagnetic ordering or some other phase transition is occurring at around 170 K. The occurrence of the superconducting state correlates well with the disappearance of the high-temperature peak in the susceptibility. The similar correlation is observed in the Cr-Re system.[4] In Cr-Re alloys the zero-field nuclear magnetic resonance of the Re nuclei shows that antiferromagnetic order remains in the concentration region higher than 17 at.% Re and coexists with superconductivity.[4,7] The susceptibility peak observed in the Cr-Ru alloys at around 170 K is clear and sharp compared with that in the Cr-Re alloys. Therefore, it is expected that superconductivity and itinerant-electron antiferromagnetism coexist also in the bcc Cr-Ru phase.

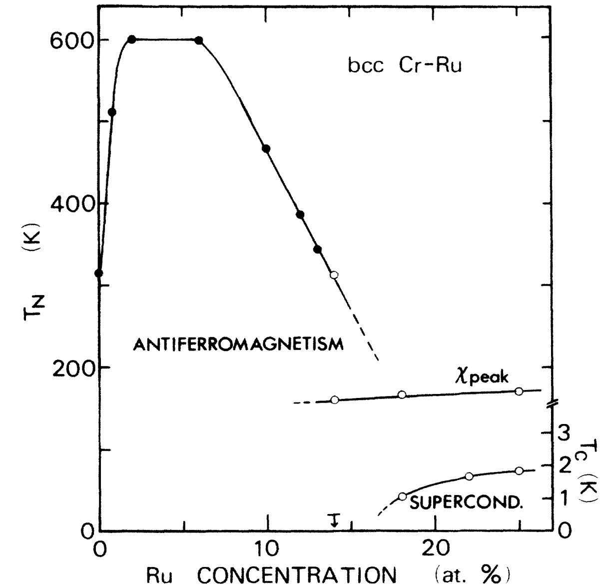

FIG. 6. Superconducting and magnetic transition temperatures of bcc Cr-Ru alloys. The closed circles are the magnetic transition temperatures from Refs. 2 and 6.

Recently, Damaschke and Felsch[8] have made an analysis of the upper critical field of Cr-Re alloys and reported that the critical-field curves are adequately described by the theory for dirty superconductors. Moreover, they have pointed out that all the experimental data of Cr-Re alloys have a drawback coming from sample inhomogeneities. The width of the superconducting transition in the Cr-Ru alloys is ten times smaller than that in the Cr-Re alloys. Thus the Cr-Ru system will provide more accurate information than the Cr-Re system on the superconducting properties in the state where it coexists with itinerant-electron antiferromagnetism.

In the Cr-Ru alloys the superconducting transition temperature T_c^R determined from the resistivity is higher than the T_c determined from the specific heat. The magnetization measurement shows that the lower critical field is less than 10 Oe between T_c^R and T_c and increases rapidly below T_c with decreasing temperature. The difference between T_c^R and T_c is 0.3–0.6 K. However, the transition width estimated by both measurements is $\sim$0.05 K. In general, two transition temperatures would be observed when the sample is inhomogeneous. In an inhomogeneous solid solution, the transition width is relatively broad and the temperature dependence of the upper critical field has an upward curvature near T_c.[9,10] The transition width and the upper critical field of the Cr-Ru system have none of the tendencies associated with this type of inhomogeneous superconductor. Therefore, the resistive transitions T_c^R could be dominated by a low-concentration impurity phase, while the specific heat T_c could represent the majority bulk solid solution. The superconductivity between T_c^R and T_c might result from the surface superconducting state or a new state related to itinerant-electron antiferromagnetism. More detailed experiments are required in order to understand the origin of the superconductivity between T_c^R and T_c and to confirm itinerant-electron antiferromagnetism below ~ 170 K.

IV. CONCLUSION

Cr-Ru alloys with bcc structure exhibit superconductivity below ~ 2 K in the concentration region higher than ~ 17 at.% Ru. Since the magnetic susceptibility of these alloys shows a peak around 170 K, it is expected that itinerant-electron antiferromagnetism coexists with superconductivity as in the Cr-Re system.

[1]B. T. Matthias, T. H. Geballe, V. B. Compton, E. Corenzwit, and G. W. Hull, Jr., Phys. Rev. **128**, 588 (1962).
[2]J. G. Booth, Phys. Status Solidi **7**, K157 (1964).
[3]J. Muheim and J. Müller, Phys. Kondens. Mater. **2**, 377 (1964).
[4]Y. Nishihara, Y. Yamaguchi, T. Kohara, and M. Tokumoto, Phys. Rev. B **31**, 5775 (1985).
[5]K. Takeda and T. Kohara, Phys. Lett. **106A**, 321 (1984).
[6]K. Fukamichi, Trans. Jpn. Inst. Met. **18**, 81 (1977).
[7]T. Kohara, K. Asayama, Y. Nishihara, and Y. Yamaguchi, Solid State Commun. **49**, 31 (1984).
[8]B. Damaschke and W. Felsch, Z. Phys. B **63**, 179 (1986).
[9]J. D. Klump, M. P. Zaitlin, and J. D. Verhoeven, Phys. Rev. B **26**, 3639 (1982).
[10]L. D. Woolf, S. E. Lambert, M. B. Maple, F. Acker, H. C. Ku, W. Odoni, and H. R. Ott, J. Low Temp. Phys. **51**, 117 (1983).