PHYSICAL REVIEW B VOLUME 25, NUMBER 9 1 MAY 1982

Magnetic order and superconductivity in $Ho(Ir_xRh_{1-x})_4B_4$ compounds

F. Acker
Institut de Physique Expérimentale, Université de Lausanne, 1015 Lausanne, Switzerland

H. C. Ku*
Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, California 92093

(Received 19 November 1981)

Magnetization data for $Ho(Ir_xRh_{1-x})_4B_4$ compounds in the temperature range 1.4 to 4.2 K and in fields up to 68 kOe are presented. Evidence of antiferromagnetic order below $T_m \simeq 1.75$ K, is found for $x = 0.5$. This modifies the phase diagram drawn previously and suggests that the boundary $T_m(x)$ might be continuous for $0 \leq x \leq 1$. The order seems to be complex and to change from ferromagneticlike to antiferromagneticlike near $x = 0.17$, the only concentration at which χ is isotropic (an "easy" axis at $x = 0$ becomes "hard" for $x > 0.17$). Antiferromagnetic interactions between ions appear to play a role at all concentrations. The higher is x, the larger is the difference between the ordering temperature T_m and the Curie-Weiss intercept Θ. In $Ho(Ir_{0.17}Rh_{0.83})_4B_4$, T_m still coincides, within ±0.05 K, with the lower superconducting critical temperature, T_{c2}, and the value of the upper critical field, H_{c2}, drops, by cooling, in a narrow interval above T_{c2}. For the antiferromagnetic $Ho(Ir_{0.7}Rh_{0.3})_4B_4$ compound ($T_m = 2.7$ K) spin flop or distortion of the magnetic structure is detectable, at 1.4 K, for low values of the external field ($H > 2$ kOe).

I. INTRODUCTION

The study of magnetic superconductors was given a new impetus when it was reported that superconductivity is destroyed at the onset of magnetic order in the compounds $HoMo_6S_8$ (Ref. 1) and $ErRh_4B_4$ (Ref. 2). Investigations, frequently emphasizing a possible "interplay" between superconductivity and magnetism, soon extended to related pseudoternary systems of compounds containing two different rare earths.[3] The partial substitution of the transition metal has been less frequently tried. Compounds of this type are, however, especially interesting from the point of view of magnetism, because the rare-earth lattice remains intact (lattice parameters may vary, but the occupation of the rare-earth sites does not).

Starting from the magnetic compound $HoRh_4B_4$, the gradual substitution of Rh by Ir was shown to cause superconductivity to appear for $x = 0.07$ in $Ho(Ir_xRh_{1-x})_4B_4$, and to turn the magnetic order into antiferromagnetism at higher concentration.[4] The unusual feature of antiferromagnetic ordering above the superconducting transition temperature was recently confirmed by heat-capacity and thermal conductivity experiments on $Ho(Ir_{0.7}Rh_{0.3})_4B_4$ (Ref. 5).

Several questions about the system $Ho(Ir_xRh_{1-x})_4B_4$ were left open and motivated the present work. The magnetic phase diagram we published[4] was incomplete and somewhat misleading, with no indication of an ordering temperature for $0.22 < x < 0.5$. Since all the compounds crystallize with the same structure[4] one may expect the magnetic transition line $T_m(x)$ to be continuous. A question immediately arises: What is the exact nature of the magnetic order? If the order is qualitatively dissimilar for $x \simeq 0$ (e.g., ferromagnetic) and for $x \simeq 0.7$ (antiferromagnetic) the critical concentration at which its nature changes has to be determined. Will there be a Lifshitz point?[6] Another possibility is a transition between two different antiferromagnetic structures (in the general meaning). Indeed, our magnetic data for $HoRh_4B_4$ are compatible with a structure which, although locally ferromagneticlike, has no resultant magnetic moment in the bulk, like sinusoidal antiferromagnetism with a long period.[7] It is thus even conceivable that the order does not change qualitatively when x increases.

The information about the destruction of superconductivity by magnetic ordering in the present system is also incomplete. If the boundary $T_m(x)$ is a continuous curve on the phase diagram, there will necessarily be a segment $T_{c2}(x)$ distinct from $T_m(x)$ and joining the x axis, provided that superconductivity persists down to $T = 0$ in the antiferromagnetic compounds. If the magnetic transitions are sharp one expects to find always $T_{c2} \leq T_m$. Curiously, $T_{c2} > T_m$ was recently reported[8] for Ho-rich $(Er_{1-x}Ho_x)Rh_4B_4$ compounds. This lead us to try to determine T_m by a more refined method for the system $Ho(Ir_xRh_{1-x})_4B_4$.

II. EXPERIMENTAL

The samples were prepared by arc melting followed by annealing.[4] Depending on the needs we used spherical (about 100 mg) or needle-shaped (down to 6.5 mg) specimen. Measurements were also performed on samples of irregular shape and on powder. The demagnetization factor plays no role in the present determination of T_m. However, its knowledge may be helpful to extract further information from the data.

The magnetization M was measured as a function of field, H, up to 68 kOe, using a moving sample magnetometer.[9] Temperature was measured by means of the vapor pressure of He and was held fixed within ±0.01 K.

As previously explained,[4,7,10] the samples show directed crystalline growth along a direction perpendicular to the surface which was in contact with the copper hearth of the arc furnace. In what follows, the magnetization measured with the field parallel or perpendicular to the cooling axis is denoted $M_{\parallel}$ and $M_{\perp}$, respectively.

III. RESULTS

Magnetic isotherms for $Ho(Ir_xRh_{1-x})_4B_4$ compounds are shown in the form of Arrott plots (M^2 vs H/M) in Figs. 1–4 for $x = 0$, 0.17, 0.50, and 0.70. Also shown, for $x = 0.17$ and for $x = 0.50$, is the temperature variation of the zero-field inverse susceptibility, as determined in the most reliable way by extrapolating the rectilinear parts of the Arrott plots to $M^2 = 0$.

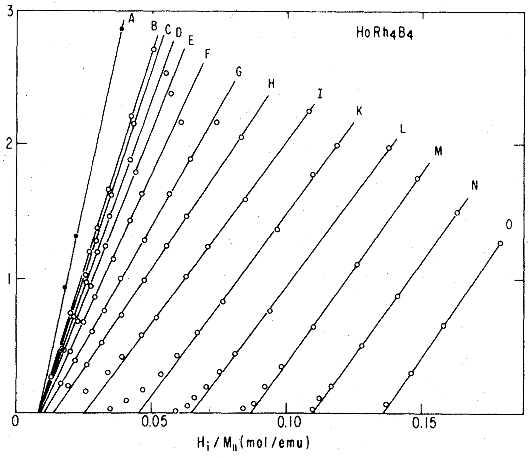

FIG. 1. Arrott plots ($M_{\parallel}^2$ vs $H/M_{\parallel}$) of the magnetization of $HoRh_4B_4$ at various temperatures near T_m after correction for demagnetization. *A*, 1.68; *B*, 4.35; *C*, 4.62; *D*, 4.92; *E*, 5.15; *F*, 5.45; *G*, 5.77; *H*, 6.03; *I*, 6.30; *K*, 6.59; *L*, 6.87; *M*, 7.15; *N*, 7.49; *O*, 7.85 K. The isotherm at 1.68 K was measured separately (from Ref. 7).

Detailed magnetic measurements of $M_{\parallel}$ for $HoRh_4B_4$ were reported by Acker and Ku (Ref. 7), from which Fig. 1 is taken. The absence of a spontaneous magnetization below the magnetic transition temperature, $T_m = 6.3$ K, is evident (the data were corrected for demagnetization). For $T < T_m$ all the Arrott plots cut the H/M axis at the same point, defining $\chi_{\min}^{-1} > 0$. Hence $M \rightarrow 0$ for $H \rightarrow 0$. Above T_m, $\chi_{\parallel}^{-1}$ follows accurately the Curie-Weiss law (with $\mu_{\text{eff}} = 10.6\mu_B$/Ho ion,[7,8] i.e., the free-ion value), whereas $\chi_{\perp}^{-1}$ does not.[7]

Figure 2(a) shows Arrott plots for $Ho(Ir_{0.17}Rh_{0.83})_4B_4$. It is qualitatively similar to Fig. 1. At each temperature the data were taken above the upper critical field H_{c2}. Below 2.18 K, the slope of the plots begins to increase and the extrapolated zero-field value of H/M becomes nearly constant, as shown in Fig. 2(b). Above 2.2 K, the Curie-Weiss law is well followed, with $\mu_{\text{eff}} = 9.75\mu_B$/Ho ion. By analogy with $HoRh_4B_4$, T_m is defined here as the temperature at which $\chi^{-1}(T)$ *deviates* from the Curie-Weiss law (see Ref. 7). In Fig. 2(b) the arrow indicates the value of T_{c2} found by ac susceptibility

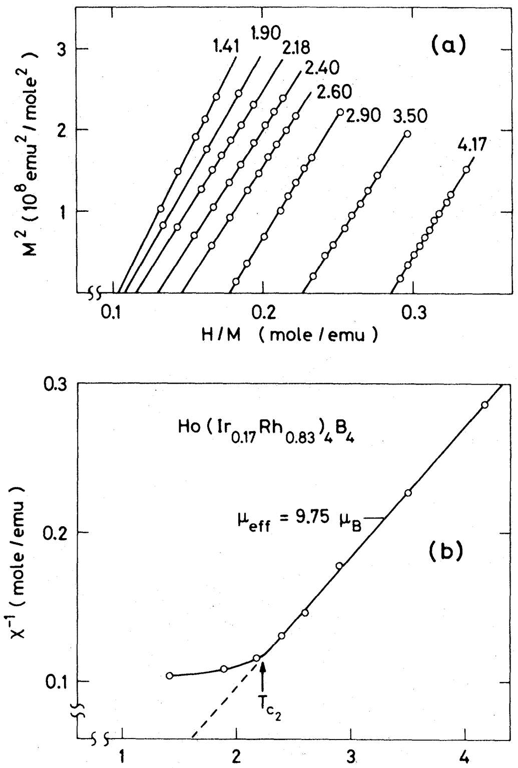

FIG. 2. (a) Arrott plots (M^2 vs H/M) of the magnetization of a $Ho(Ir_{0.17}Rh_{0.83})_4B_4$ powdered sample at various temperatures between 1.41 and 4.17 K (not corrected for demagnetization). (b) Variation with temperature of the extrapolated zero-field inverse susceptibility, obtained from least-squares fits of (M^2 vs H/M) data to straight lines.

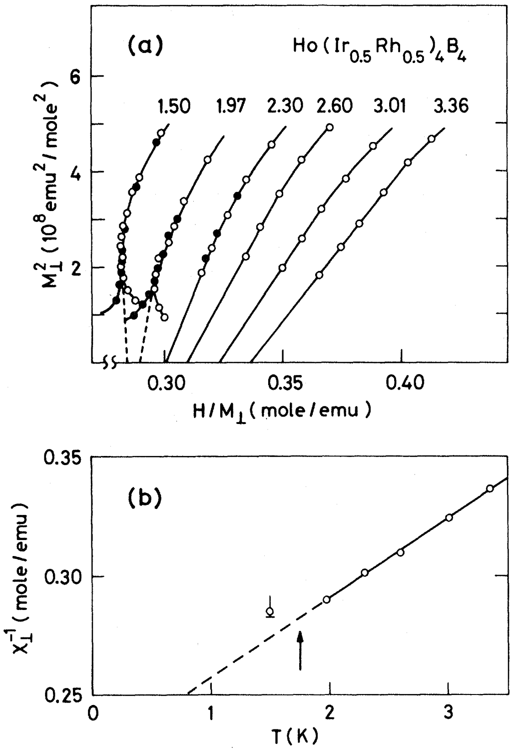

FIG. 3. (a) Arrott plots ($M_\perp^2$ vs $H/M_\perp$) of the magnetization of $Ho(Ir_{0.5}Rh_{0.5})_4B_4$ at various temperatures between 1.50 and 3.36 K, for H increasing (○) and H decreasing (●) (not corrected for demagnetization). (b) Variation with temperature of the zero-field inverse susceptibility, obtained from the nearly rectilinear parts of the Arrott plots (see text).

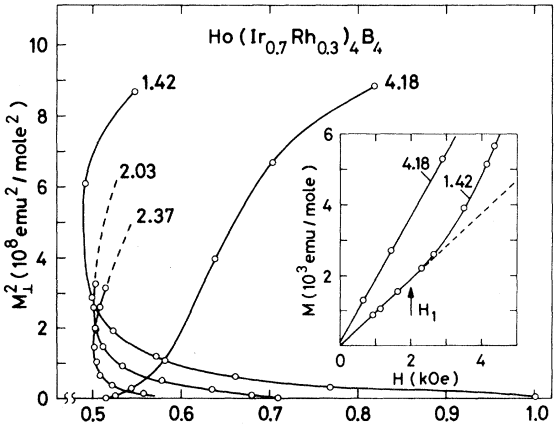

FIG. 4. Arrott plots ($M_\perp^2$ vs $H/M_\perp$) of the magnetization of $Ho(Ir_{0.7}Rh_{0.3})_4B_4$ at various temperatures between 1.42 and 4.18 K (not corrected for demagnetization). Insert, low-field magnetization curves (M vs H) at 1.42 and 4.18 K.

on the same sample.[4] It comes out that, within ±0.05 K, $T_m = T_{c2} = 2.23$ K. The data in Fig. 2 are not corrected for demagnetization (powdered sample) so that the value of the initial susceptibility at T_m cannot be determined. Complementary measurements on a spherical sample ($T_m = 2.30$ K) show that $\chi(T < T_m)$ for $x = 0.17$ is finite (≈ 17 emu/mole) and about six times smaller than in $HoRh_4B_4$. Hence the measured isotherms do not cut the M^2 axis and the absence of a spontaneous magnetization for Rh-rich $Ho(Ir_xRh_{1-x})_4B_r$ compounds is confirmed.

The look of the Arrott plots at low temperature for $x = 0.5$ and 0.7 ($M_\perp$, Figs. 3 and 4) contrasts with that of Figs. 1 and 2. Since the antiferromagnetic compound $Ho(Ir_{0.7}Rh_{0.3})_4B_4$ is not superconducting above 1.6 K, the measurements shown in Fig. 4 are not affected by supercurrents. Below $T_N = 2.7$ K, the Arrott plots for moderate fields are retrograde, curved, and reversible. In other words the susceptibility M/H increases with increasing field (by about 125%, at 1.42 K, taking into account a correction for demagnetization). A similar tendency is discernible in Fig. 3(a), for $Ho(Ir_{0.5}Rh_{0.5})_4B_4$, although the low-field part of the curves ($H < H_{c2}$) is missing. Data points taken below H_{c2}, for H increasing and decreasing, are shown for $T = 1.50$ and 1.97 K. The Arrott plot for $T = 1.50$ K is slightly retrograde, just above H_{c2}. Extrapolation to $H = 0$ of the data in this range of field (dashed lines) gives a lower limit for χ^{-1} at 1.50 and 1.97 K. Even so, the inverse susceptibility [Fig. 3(b)] clearly deviates from the Curie-Weiss law between 1.5 and 2 K. We estimate that $T_m = 1.75 \pm 0.15$ K. From the analogy with the magnetic behavior of $Ho(Ir_{0.7}Rh_{0.3})_4B_4$ we conclude that the order has an antiferromagnetic character for $x = 0.5$. This is confirmed by the relative smallness of χ at T_m. The slope of the Curie-Weiss plot at 3 K corresponds to an effective moment of $18\mu_B$/Ho, which results from the presence of traces ($\approx 1\%$) of an impurity phase with $T_m \simeq 18$ K (Ref. 11), quite probably $Ho(Ir_xRh_{1-x})_3B_2$ (Ref. 12).

Coming back to $Ho(Ir_{0.7}Rh_{0.3})_4B_4$, we see in the insert of Fig. 4 that M varies first linearly with H, at 1.42 K, up to a threshold field $H_1 \simeq 2$ kOe. For higher fields M/H increases and goes through a maximum at $H \simeq 11$ kOe, whereas the differential susceptibility dM/dH peaks at $H_{max} \simeq 6$ kOe (not shown). Both H_1 and H_{max} decrease when T increases, and vanish at T_N (Néel temperature).

In order to gain additional information about the evolution of the magnetic order in the present system we looked at the anisotropy of the magnetic properties in high fields. Anisotropy should be more striking in the low-field susceptibility, but $\chi(H \simeq 0)$ is strongly affected by the presence of traces of impurity phases and by the sample geometry (demagnetization effect). In high fields these two effects become practically negligible. However, the magnetic structure

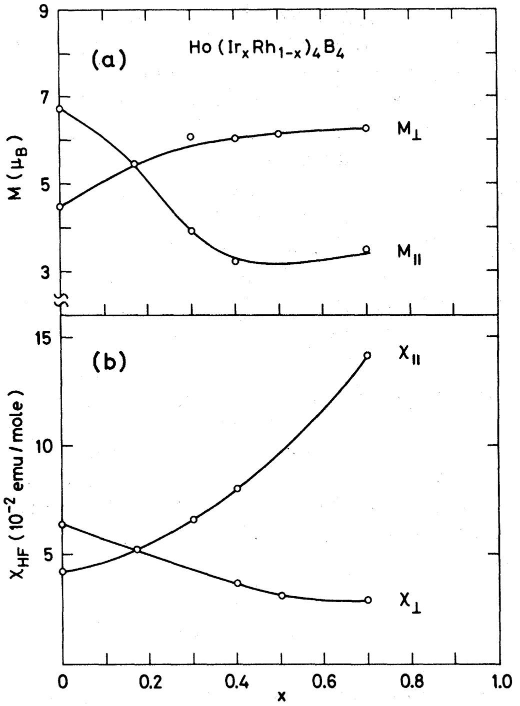

FIG. 5. Variation with Ir concentration of (a) the magnetization $M_{\parallel}$, respectively $M_{\perp}$, of Ho$(\mathrm{Ir}_x\mathrm{Rh}_{1-x})_4\mathrm{B}_4$ samples, measured in a field of 68 kOe applied in two directions (see text); (b) the corresponding differential susceptibility for $H=60$ kOe. All data were taken at 4.2 K.

may be strongly distorted by the field.

Figure 5 shows high-field data for Ho$(\mathrm{Ir}_x\mathrm{Rh}_{1-x})_4\mathrm{B}_4$ compounds at 4.2 K, with the applied field parallel or perpendicular to the cooling axis. The magnetization in a field of 68 kOe is shown in Fig. 5(a) and the differential susceptibility, $\chi_{\mathrm{HF}} = dM/dH$ for $H=60$ kOe, in Fig. 5(b). It is readily seen that the anisotropy vanishes (and "changes sign") at $x \simeq 0.17$ and that the high-field susceptibility is larger where the high-field magnetization is smaller. At 68 kOe, the magnetization of all the samples studied is smaller than $7\mu_B$ per Ho atom, even with the field in the "easy" direction. In fields higher than 30 kOe the value of $M_{\perp}$ for the "antiferromagnetic" compound with $x=0.7$ is practically equal to that of $M_{\parallel}$ for the "ferromagnetic" HoRh$_4$B$_4$.

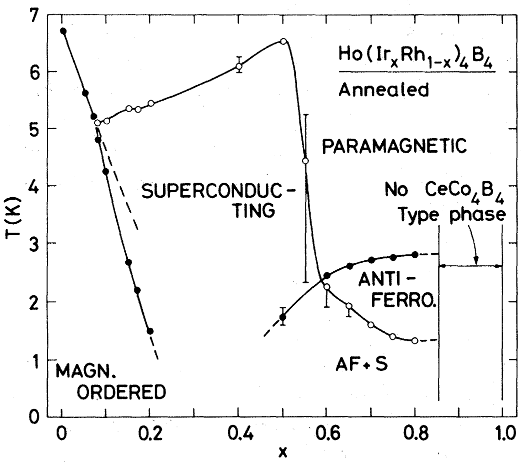

FIG. 6. Low-temperature phase diagram for the system Ho$(\mathrm{Ir}_x\mathrm{Rh}_{1-x})_4\mathrm{B}_4$ determined on annealed samples. The concentration of the CeCo$_4$B$_4$-type phase decreases sharply only above $x=0.8$.

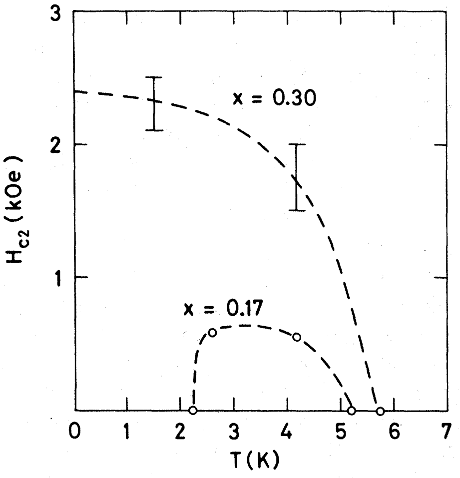

FIG. 7. Preliminary data for the variation with temperature of the upper critical field H_{c2} of two Ho$(\mathrm{Ir}_x\mathrm{Rh}_{1-x})_4\mathrm{B}_4$ compounds ($x=0.17$ and 0.30).

Shown in Fig. 6 is a modified but still incomplete magnetic and superconducting phase diagram for the system Ho$(\mathrm{Ir}_x\mathrm{Rh}_{1-x})_4\mathrm{B}_4$, based on the results of Ref. 4. We corrected the T_m vs x boundary between $x=0.5$ and 0.6. On the Rh-rich side, the present work establishes that, within ±0.05 K, $T_m = T_{c2}$ for $x=0.17$. The results shown in Fig. 5 indicate that the boundary $T_m(x)$ should become distinct from $T_{c2}(x)$ above $x \simeq 0.17$. Whether T_{c2} decreases to zero in the way suggested in Ref. 4 is uncertain.

Preliminary data for $H_{c2}(T)$, defined as the field (corrected for demagnetization) above which the $M(H)$ curve is reversible, are shown in Fig. 7, for $x=0.17$ and 0.30. For comparison, $H_{c2}(T \rightarrow 0) \simeq 6$ kOe in Ho$(\mathrm{Ir}_{0.7}\mathrm{Rh}_{0.3})_4\mathrm{B}_4$ (from χ_{ac} measurements, Ref. 5). For $x=0.17$, the drop of H_{c2} when T is reduced from above $T_{c2} = T_m$ is anomalously sharp, as observed in Ho-rich $(\mathrm{Er}_{1-x}\mathrm{Ho}_x)\mathrm{Rh}_4\mathrm{B}_4$ compounds.[8,13]

IV. DISCUSSION

Our main concern in this work was to determine the magnetic transition temperature, T_m. Judging from the result, that $T_m = T_{c2}$ for $x=0.17$, the method used appears correct. Indeed, for Ho-rich

$(Er_{1-x}Ho_x)Rh_4B_4$ compounds, the magnetic transition temperature found by heat-capacity measurements is identical to T_{c2} found by ac susceptibility[14] (at most $T_{c2} \leq T_m$). The nature of the magnetic order of the Ho^{3+} ions in that system and in Rh-rich $Ho(Ir_xRh_{1-x})_4B_4$ is probably similar, since $HoRh_4B_4$ is the end member of both systems. Hence the comparison between them is meaningful.

The reason why Adrian *et al.*[8] find T_m as far as 1.3 K below T_{c2} in $(Er_{0.4}Ho_{0.6})Rh_4B_4$ is, we believe, simple and revealing. These authors extrapolate the Curie-Weiss plot, χ^{-1} vs T, down to $\chi^{-1}=0$, defining the Curie-Weiss temperature Θ. Quite probably, the true zero-field susceptibility of the compounds in question remains finite, for $T \leq T_m$, as found here for $Ho(Ir_xRh_{1-x})_4B_4$ compounds (see also Ref. 7). Hence the magnetic transition occurs at a temperature T_m significantly higher than Θ. More precisely it appears that $(T_m - \Theta)$ increases when x increases from zero in $Ho(Ir_xRh_{1-x})_4B_4$ and $(Ho_{1-x}Er_x)Rh_4B_4$ systems, which could mean that antiferromagnetic interactions become stronger. This interpretation is suggested by the occurrence of antiferromagnetism in the first system and might be not appropriate to the second. However, anisotropy in the magnetic properties indicates that antiferromagnetic interactions already play a role in $HoRh_4B_4$ and cause the magnetic order to be complex.[7] To our knowledge the refined, low-angle neutron-diffraction experiments necessary to detect a magnetic structure with a long period were not undertaken for $HoRh_4B_4$. Such measurements were carried out with success for $ErRh_4B_4$ (Ref. 15) and $HoMo_6S_8$ (Ref. 16) where periods as long as 200 Å were found. It would be very interesting to check if any kind of modulated structure also appears in $HoRh_4B_4$, because this compound is not superconducting, unlike $ErRh_4B_4$ and $HoMo_6S_8$. The occurrence of these spatial variations of magnetization were precisely interpreted as due to an interaction between superconductivity and magnetic order.[15,16] A negative result of similar neutron-diffraction experiments for $HoRh_4B_4$ would strengthen that interpretation, whereas a positive result would be in line with our magnetization measurements.

Coming back to the system $Ho(Ir_xRh_{1-x})_4B_4$, the magnetic transition at $T_N \simeq 1.75$ K we detected for $x=0.5$, together with the results for the anisotropy of the magnetization (Fig. 5), suggest that the order has an antiferromagnetic character down to $x \simeq 0.2$. The two branches of T_m vs x, in the phase diagram (Fig. 6) might join there. This resembles the situation found recently in (rare-earth) $(Ru_xRh_{1-x})_4B_4$ compounds (body-centered tetragonal).[17,18] It has been pointed out that the destruction of superconductivity in $Dy(Ru_{0.327}Rh_{0.673})_4B_4$ could be caused by the antiferromagnetic ordering of the rare-earth ions.[17] This novel phenomenon might take place in $Ho(Ir_xRh_{1-x})_4B_4$ as well, for x just above 0.2 (or even below, if our views about the magnetic order in $HoRh_4B_4$ are qualitatively correct).

The application of a magnetic field strongly affects the magnetic structure of the compound $Ho(Ir_{0.7}Rh_{0.3})_4B_4$. Its magnetization $M_\perp$, at $T=1.5$ K and $H=1$ kOe is about 20 times smaller than $M_\parallel$ for $HoRh_4B_4$ in the same conditions. As stated above, this ratio tends towards 1 for $H > 30$ kOe. The low value of the threshold field[19] ($H_1 \simeq 2$ kOe at 1.42 K) above which the $M(H)$ curve becomes concave upwards may indicate that the antiferromagnetic interactions between ions are weak. Ferromagnetic interactions probably also play a role, leading to a complex magnetic structure. The moment of the Ho^{3+} ions is probably oriented along the tetragonal c axis for all values of x (Ref. 10).

V. CONCLUSION

Clearly, all the questions asked in the introduction cannot be solved with only the help of bulk magnetization measurements above 1.4 K, and further work on the system $Ho(Ir_xRh_{1-x})_4B_4$ will be necessary in particular low-temperature heat-capacity experiments.

The present results are consistent with a continuous variation of the magnetic transition temperature with Ir concentration. Special attention should be directed to the compounds in the range $0.20 < x < 0.40$ where the $T_{c2}(x)$ boundary in the phase diagram must become distinct from $T_m(x)$, if the latter is continuous. One may imagine interesting situations if $T_{c2}(x)$ is not vertical in this region, namely, a return to the normal state at a temperature well below T_m. For $x=0.17$ we found $T_m = T_{c2}$.

The exact nature of the magnetic order and its evolution with Ir concentration is still unknown. Detailed neutron-diffraction experiments would help to clarify this point and could bring new information relevant to a wider class of ternary compounds where superconductivity is affected by the magnetic ordering of rare-earth ions.

ACKNOWLEDGMENTS

We wish to thank G. H. Lander, S. K. Sinha, and S. E. Lambert for helpful discussions. This research was supported by the Swiss National Science Foundation and by the U.S. National Science Foundation under Grant No. DMR80-17723.

[*] Present address: Department of Physics, National Tsing Hua University, Hsinchu, Taiwan, Republic of China.

[1] M. Ishikawa and Ø. Fischer, Solid State Commun. 23, 37 (1977).

[2] W. A. Fertig, D. C. Johnston, L. E. DeLong, R. W. McCallum, M. B. Maple, and B. T. Matthias, Phys. Rev. Lett. 38, 987 (1977).

[3] See, for example, *Proceedings of the International Conference on Ternary Superconductors, Lake Geneva, Wisconsin, September 24–26, 1980,* edited by G. K. Shenoy (North-Holland, Amsterdam, 1981).

[4] H. C. Ku, F. Acker, and B. T. Matthias, Phys. Lett. 76A, 399 (1980).

[5] L. D. Woolf, S. E. Lambert, M. B. Maple, H. C. Ku, W. Odoni, and H. R. Ott, Physica (Utrecht) 108B, 761 (1981).

[6] See, for example, R. M. Hornreich, M. Luban, and S. Shtrikman, Phys. Rev. Lett. 35, 1678 (1975); R. M. Hornreich, J. Magn. Magn. Mater. 15–18, 387 (1980); C. C. Becerra, Y. Shapira, N. F. Oliveira, Jr., and T. S. Chang, Phys. Rev. Lett. 44, 1692 (1980).

[7] F. Acker, and H. C. Ku, J. Low Temp. Phys. 42, 449 (1981); see also F. Acker and H. C. Ku, J. Magn. Magn. Mater. 24, 47 (1981).

[8] H. Adrian, K. Müller, and G. Saemann-Ischenko, Phys. Rev. B 22, 4424 (1980).

[9] F. Acker and R. Huguenin, J. Magn. Magn. Mater. 12, 58 (1979).

[10] H. C. Ku, H. F. Braun, and F. Acker, Physica (Utrecht) 108B, 1231 (1981).

[11] F. Acker, unpublished susceptibility data. The concentration of the impurity phase appears to be maximum for x in the range 0.5–0.6. For $x = 0.7$ the effect of a magnetic impurity phase on the susceptibility is hardly detectable. There is possibly a link between these facts and the sudden change in the composition variation of the lattice parameter c found around $x = 0.6$ for two (rare-earth) $(Ir_xRh_{1-x})_4B_4$ systems. See H. C. Ku and F. Acker, Solid State Commun. 35, 937 (1981).

[12] H. C. Ku and G. P. Meisner, J. Less-Common Met. 78, 99 (1981).

[13] M. Ishikawa, Phys. Lett. 74A, 263 (1979).

[14] H. B. MacKay, L. D. Woolf, M. B. Maple, and D. C. Johnston, Phys. Rev. Lett. 42, 918 (1979).

[15] D. E. Moncton, D. B. McWhan, P. H. Schmidt, G. Shirane, W. Thomlinson, M. B. Maple, H. B. MacKay, L. D. Woolf, Z. Fisk, and D. C. Johnston, Phys. Rev. Lett. 45, 2060 (1980).

[16] J. W. Lynn, G. Shirane, W. Thomlinson, and R. N. Shelton, Phys. Rev. Lett. 46, 368 (1981); J. W. Lynn, A. Ragazzoni, R. Pynn, and J. Joffrin, J. Phys. (Paris) Lett. 42, L45 (1981).

[17] H. C. Hamaker and M. B. Maple, Physica (Utrecht) 108B, 757 (1981).

[18] H. Iwasaki, M. Isino, and Y. Muto, Physica (Utrecht) 108B, 759 (1981).

[19] The present definition of a threshold field H_1 is loose. Within experimental error the low-field data can also be fitted to $H = AM + BM^3$, according to which $M(H)$ is slightly curved down to $H = 0$. The field $H_{\max}$ could be taken as the critical field for the "breakdown" of the zero-field magnetic structure. The points $H_{\max}(T)$ must fall nearly, but not exactly, on the curve $T_N(H)$ (apparent magnetic transition temperature) defined in Ref. 5 by the maxima of $\chi_{ac}(T)$ in various fields.