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Destruction of Ferromagnetism in $ZrZn_2$ at High Pressure

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The variation of the Curie temperature T_c for the weak ferromagnet $ZrZn_2$ has been determined as a function of hydrostatic pressure up to 25 kbar. T_c is found initially to decrease linearly with pressure at a rate $\partial T_c/\partial P = (-1.8 \pm 0.04) \times 10^{-3}\,{}^\circ\mathrm{K\ bar}^{-1}$, followed by an abrupt change of slope at 7.5 kbar and a rapid fall, extrapolating to a critical pressure P_c of 8.5 kbar for the destruction of ferromagnetism. The measured $T_c(P)$ curve agrees well with the relationship $T_c(P) = T_c(0)(1 - P/P_c)^{1/2}$ which is derived from the theory of itinerant ferromagnetism.

The Laves phase compound $ZrZn_2$ is considered to be a prime example of a weak itinerant electron ferromagnet.[1] This compound is composed of two elemental superconductors, neither of which possess the usual electronic configurations (3*d*, 4*f*, or 5*f*) found in magnetic materials. A small effective magnetic moment ($\sim 0.15\ \mu_B$ per Zr atom) and a low Curie temperature ($\sim 20^\circ$K) point towards the itinerant nature of this compound.[2] It is also expected for itinerant electron systems that large pressure effects will be observed in the magnetic properties.[3] Indeed, our measurements of the Curie temperature of $ZrZn_2$ indicate the destruction of the ferromagnetic state for pressures in excess of 8.5 kbar. This represents the first known example of a ferromagnetic system in which the magnetic behavior can be, apparently, completely suppressed by the application of pressure.

The sample of $ZrZn_2$ consisted of three pieces (total mass ~ 140 mg) broken from a dense conglomerate of large crystallites (typical dimensions ~ 0.1 cm) which had been grown[4] by the vapor transport technique. An x-ray examination[5] indicated that the material is single phase and has the C15, Laves phase structure with a lattice parameter of 7.396 ± 0.001 Å. The onset of the ferromagnetic state was detected by means of an ac mutual-inductance technique in which the off-balance signal from a pair of matched and opposed secondaries, one of which surrounds the sample, is continuously monitored as a function of temperature. The applied 150-Hz signal is estimated to produce a peak-to-peak field at the sample of ~ 0.2 G. The inductance measurements were made in a clamp device, in which pressure applied at room temperature using a hydrostatic pressure medium of a 1:1 mixture of *n*-pentane and isoamyl alcohol may be contained, permitting the removal of the high-pressure stage from the press, and its transfer to a cryostat in which the lowest attainable temperature was 1°K. The pressure at the low temperature was determined relative to the shift in the superconducting transition temperature of lead.[6]

The observed transition curves are similar in appearance to, though somewhat sharper than, those recorded by Wayne and Edwards[7] using a similar detection technique in their study of the pressure dependence of T_c to 3 kbar for a powdered sample of $ZrZn_2$. Such curves are a measure of the initial susceptibility and are usually not very suitable for an accurate determination of T_c. However, provided that the overall shape of the curve does not change significantly under pressure, the relative shift of these curves does allow a reliable measure of the change of T_c with pressure. We have elected to use the intersection of the line drawn through the steeply arising central portion of the transition curve and the extrapolation of the background trace to define an effective T_c. Justification for this procedure is found in the good agreement between the present value for the initial pressure dependence of T_c and that $[(-1.95\pm0.1)\times10^{-3}\,°\mathrm{K\ bar}^{-1}]$ determined by Wayne and Edwards.[7] They found that the zero-pressure T_c of their sample, derived from a conventional plot of M^2 vs H/M following measurements of the magnetization M in a magnetic field H, corresponded to a temperature approximately one third of the way through the transition curve determined from the inductance measurement, and they subsequently determined the pressure dependence of T_c relative to the shift of this point.

Our data for the variation of T_c with pressure, which are presented in the lower portion of Fig. 1, were taken during the course of a number of cycles of increasing and decreasing pressure up to a maximum pressure of 25 kbar. The line drawn through the data points represents the expression

$$T_c = (22.2 - 1.8P)°\mathrm{K}, \qquad (1)$$

with P in kbar. An extrapolation of this initial variation of T_c with pressure would suggest that the ferromagnetism could be destroyed for pressures in excess of ~12 kbar. However, at 7.5 kbar there is an abrupt change of slope, and T_c rapidly falls and extrapolates to zero at about 8.5 kbar.

The magnitude of the signal associated with the ferromagnetic transition was found to decrease as the pressure increased; and in the upper portion of Fig. 1 we show the ratio of the signal, relative to the zero-pressure transition, as a function of pressure. We see that the signal ratio decreases linearly with pressure and also extrapolates to zero at 8.5 kbar. Although it is tempting to equate the magnitude of the signal with the magnetization, it must be remembered that this is proportional to the initial susceptibility, and therefore cannot be simply related to the saturation magnetization. The loss of signal placed a serious limitation on the determination of T_c close to the critical pressure since variations in the background signal prevented the unambiguous resolution of transitions with a signal ratio less than $\sim10^{-2}$. The solid symbol in Fig. 1 represents a measurement which was made close to the limiting signal level and is therefore considered to be rather less reliable than the other data points. No magnetic transitions above 1.0°K were detected at pressures of 8.8, 10.5, 16.1, and 24.4 kbar. In addition, since the initial susceptibility measurement is also sensitive to the occurrence of superconductivity, it may also be noted that no superconducting transition occurred over this range of pressure and temperature.

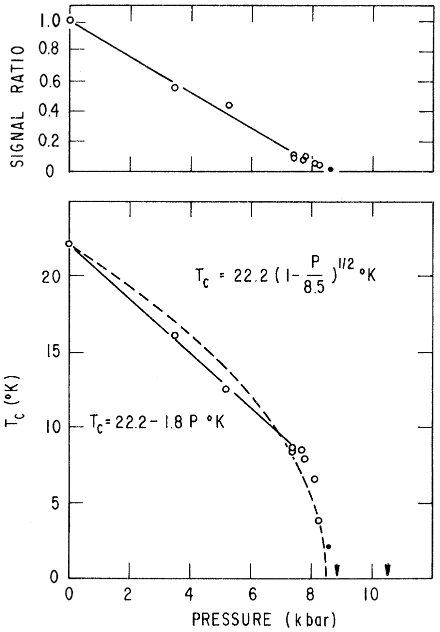

FIG. 1. Upper portion: variation of the magnitude of the signal corresponding to the transition to the ferromagnetic state as a function of pressure for $ZrZn_2$. Lower portion: pressure dependence of the Curie temperature for $ZrZn_2$. The arrows represent runs made to 1.0°K for which no magnetic transition was detected. Similar runs were also made at 16.1 and 24.4 kbar. See text for the explanation of the solid symbol.

Extensive discussions of the pressure dependence of the Curie temperature for itinerant ferromagnets, including in particular $\mathrm{ZrZn_2}$, have been given by Wohlfarth[3,8] who derived the relationship

$$\partial T_c/\partial P = -A\chi T_c, \qquad (2)$$

where A is a quantity related to the forced magnetostriction and χ is the magnetic susceptibility. With the use of this relationship and the values of A derived from the magnetostriction data of Ogawa and Waki[9] and more recently Fawcett and Meincke,[10] estimates for the initial $\partial T_c/\partial P$ of -2.4×10^{-3} and -2.8×10^{-3} °K bar^{-1}, respectively, are obtained which are in reasonable agreement with the directly measured values.

Wohlfarth has also derived the more specific relationship[3,8]

$$\partial T_c/\partial P = 2\kappa CNN(E_\mathrm{F})\mu_\mathrm{B}{}^2 T_\mathrm{F}{}^2/T_c, \qquad (3)$$

where κ is the compressibility; C is a complex quantity involving the volume derivatives of the exchange interaction and the density of states, $N(E_\mathrm{F})$; N is the number of atoms per unit volume; μ_B is the Bohr magneton; and T_F is a degeneracy temperature which is defined in terms of the fine structure in the density-of-states curve. From the pressure measurements of Wayne and Bartel[11] for the Invar alloys Fe-Nd, Fe-Pd, and Fe-Pt, it is found that $\partial T_c/\partial P \approx -1.6/T_c$ °K bar^{-1}. Noting that the values of C and $N(E_\mathrm{F})$ for $\mathrm{ZrZn_2}$ are essentially the same as those for the Invar alloys, and by scaling with the appropriate value for T_F, Wohlfarth made the estimate of -1.8×10^{-3} °K bar^{-1} for the pressure dependence of $\mathrm{ZrZn_2}$. This value is in surprisingly good agreement with that observed.

In view of the success of this simplified form of (3) for the prediction of the *initial* pressure dependence, we might anticipate that

$$\partial T_c/\partial P = -\alpha/T_c \qquad (4)$$

(where α is independent of pressure) would provide an appropriate description of the entire variation of T_c with pressure. Thus, on integrating we have

$$T_c(P) = T_c(0)(1 - P/P_c)^{1/2}, \qquad (5)$$

where P_c $[=T_c{}^2(0)/2\alpha]$ is the critical pressure for the disappearance of ferromagnetism, i.e., $T_c(P_c) = 0$. The broken line in Fig. 1 represents (5) with $T_c(0) = 22.2$°K and $P_c = 8.5$ kbar. In view of the simplifying assumptions which are embodied in (5) regarding the pressure independence of the energy band structure, the fit to the data is considered to be remarkably good. Had we taken $T_c(0)$ and P_c to be arbitrary fitting parameters rather than the directly determined quantities, the fit could be improved by least-squares analysis. On the other hand, the variation of T_c for a reduction to some 0.36 of its initial value would appear to be much closer to linear than (5) would allow. It may be noted in this respect that Wayne and Bartel[11] also report that $\partial T_c/\partial P$ is constant over the pressure range 0–25 kbar of their investigation for the Invar alloys, where the reduction of T_c ranged from approximately 8 to 40%. More recently Leger and Susse-Loriers[12] have reported measurements of the pressure dependence of T_c to 50 kbar for the two Fe-Ni Invar alloys containing 35 and 29 at.% Ni. The corresponding decreases of T_c are approximately 50 and 70% of the zero-pressure value. They find a parabolic variation for T_c in agreement with (5) for the 35% alloy, but a linear dependence for the 29% alloy.

Such deviations from the form of (5) are not surprising if we examine the individual quantities in (3). Although the compressibility may be regarded as being independent of pressure, as would also be expected for C to first order, the band structure could be expected to be influenced by the lattice volume, thus implying some pressure dependence for the quantity $N(E_\mathrm{F})T_\mathrm{F}{}^2$. Nevertheless, the departure from linear behavior and the rapid drop of $T_c(P)$ suggest that the form of (5) is valid in the high-pressure region.

As a range of T_c values for $\mathrm{ZrZn_2}$ may be obtained by suitable heat treatment[13] or alloying,[14] it is of interest to investigate further the relationship between $\partial T_c/\partial P$ and the *initial* value of T_c. This work is presently in progress.

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Isospin Nonconservation in the Reaction $^{12}C(d, \alpha_2)^{10}B(1.74, T = 1)$†

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We question the recent conclusion that a simple compound-nucleus interpretation fails for this reaction.

Recently in this Journal von Ehrenstein *et al.*[1] reported six angular distributions of the isospin-forbidden α's from the reaction $^{12}C(d, \alpha_2)^{10}B(1.74, T=1)$ and a few additional cross-section measurements at $\theta_{c.m.} \approx 160°$ for $16 < E_d < 17$ MeV. They concluded, "In the light of these new data, it is clearly not sufficient to interpret the reaction in terms of simple compound-nucleus formation." Apparently the evidence persuasive for this conclusion was that "the angular distributions··· show a pronounced enhancement (up to a factor 5) at forward angles within the incident deuteron energy interval $E_d = 12.0$ to 17.0 MeV."

We find the latter statement possibly misleading in the following respects: (1) The data for $12 < E_d < 14$ MeV are mainly from Smith[2] and consist of ~50 angular distributions which above $E_d = 13.5$ MeV show *backward* (not forward) enhancement. (2) Of the six new angular distributions for $E_d \geq 14$ MeV (see Fig. 1 of Ref. 1) only three show forward enhancement ($E_d = 14.4$, 14.8, and 15.4 MeV). The others show backward enhancement or are essentially isotropic.

Smith's complete data[2] consist of ≈150 angular distributions for $7.2 < E_d \leq 14.0$ MeV. Parametrizing these in terms of resonant partial waves shows nothing inconsistent with a simple compound-nucleus description.[2] Therefore, we are skeptical that the conclusion of Ref. 1 follows from the limited (but qualitatively similar) data for $E_d > 14$ MeV.

Before discussing the question further, let us review certain features of this particular reaction which we find many readers overlook:

(A) As Ref. 3 shows, the very simple spin-parity combination, $0^+1^+ \rightarrow 0^+0^+$, gives $d\sigma/d\Omega \propto |\sum_l \alpha_l dP_l/d\theta|^2$. Hence, the cross section must always vanish at 0° and 180°. Also, the $l = 0$ partial wave is forbidden and only compound states of natural parity are allowed, i.e., $J^{\pi} = l^{(-)^l}$.

(B) Since the reaction is also isospin forbidden, only those natural-parity states which have appreciable isospin impurity can contribute: namely, $T = 0$ states with $T = 1$ impurity and $T = 1$ states with $T = 0$ impurity. To have such mixing, $T = 0$ and $T = 1$ states of the same spin-parity combination must lie within a few hundred keV of each other since the Coulomb matrix elements, H_C, are ~100 keV. Also the states should belong to different configurations.[4] Although only a very few ^{14}N compound states will contribute, these states must lie close to another state of the same spin and parity. These nearby levels of the same spin and parity (but mixed isospin) will interfere with each other, and therefore the resonant shapes are not of simple Breit-Wigner form. Multilevel resonance formulas apply even for the simplest cases.[5]

As a consequence of (A) all partial waves with $l > 1$ give angular distributions with strong maxima at forward and/or backward angles *independent of the reaction mechanism* involved. For a single isolated resonance (or for any number of overlapping resonances with the same parity) the forward and backward peaks are of course symmetric about 90° c.m. For almost all other cases involving compound nuclear resonances, the opposite-parity partial waves by interference enhance (or diminish) the forward peak(s) relative to the backward peak(s). For our $0^+1^+ \rightarrow 0^+0^+$ system the result is that the angular distributions resemble direct reactions especially if $l \geq 3$ waves resonate, e.g., see Fig. 1. These pseudo-