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**SUSCEPTIBILITY AND RESISTIVITY OF $\zeta$-PHASE ZINC-TRANSITION METAL COMPOUNDS**

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Résumé. — Les phases $\zeta$ des composés Zn$_{13}$Mn, Zn$_{13}$Fe et Zn$_{13}$Co sont bien ordonnées. La distance entre les ions de transition est $\approx 5$ A. L'ion Mn porte un moment magnétique alors que les ions Fe et Co n'en portent pas comme dans les solutions correspondantes dans Zn. En dessous de 20 °K, Zn$_{13}$Mn s'ordonne et est faiblement ferromagnétique. On observe un fort signal en R. P. E. et le facteur $g$ passe de la valeur 2.0 à 200 °K à la valeur 3.0 en dessous de 4 °K. A basse température la résistivité de Zn$_{13}$Fe et Zn$_{13}$Co varie considérablement avec la température, indiquant un phénomène de diffusion des électrons.

Abstract. — The $\zeta$-phase compounds Zn$_{13}$Mn, Zn$_{13}$Fe and Zn$_{13}$Co are crystallographically well-ordered with $\approx 5$ A between transitional ions. Mn carries a moment but Fe and Co do not, just as in the corresponding dilute solid solutions in Zn. Below 20 °K Zn$_{13}$Mn orders and is weakly ferromagnetic; a strong e.p.r. signal is observable and the $g$-factor increases from 2.0 to 200 °K to 3.0 below 4 °K. There is strong temperature-dependence of the resistivities of Zn$_{13}$Fe and Zn$_{13}$Co at low temperatures, indicative of electron-electron scattering.

The dilute transitional impurity dissolved in a free-electron-like solvent, and the pure transition metal itself, represent two limiting cases in the theory of magnetism. An intermediate situation which, by combining the localized nature of the former problem with the periodicity of the latter, should prove amenable to understanding, is provided by the ordered $\zeta$-phase compounds Zn$_{13}$X, where X can be Mn, Fe or Co. Brown [1] has shown that in these materials the transition ions are well-separated ($\approx 5$ A) and each is surrounded by a slightly distorted sphere of zinc atoms.

The high temperature susceptibility of $\zeta$-Mn fits a Curie-Weiss law (Fig. 1) with $\mu_B^0 = 1.82$ $\mu_B$, $\theta = 36$ °K and $X_0 = 0.53 \times 10^{-6}$ emu/g. It orders ferromagnetically at about 20 °K (deduced from a plot of $H/M$ versus $M^2$ [2]) with a saturation moment of only 0.3 $\mu_B$ per Mn ion (Fig. 2). Our e. p. r. experiments (at X-band on powder samples) show a strong signal below 200 °K, with $g = 2.0$ down to 150 °K, and an increasing positive $g$-shift at lower temperatures (Fig. 3); no correction for demagnetizing fields has been made, for at most it would account for 20 % of the observed shift. The susceptibilities of $\zeta$-Fe and $\zeta$-Co are small and almost temperature-independent (Fig. 1); the $\zeta$-Co samples show considerable variability, but are distinctly more paramagnetic than $\zeta$-Co, whose susceptibility is about equal to that of pure Zn ($-0.1 \times 10^{-6}$ emu/g).

The room temperature resistivities of these compounds are high; about 90, 30 and 20 $\mu$cm for $\zeta$-Mn, $\zeta$-Fe and $\zeta$-Co respectively, but our samples had quite good resistance ratios, generally between 10 and 30. Their resistivities are strongly temperature-dependent at low temperatures (Fig. 4); that of $\zeta$-Mn is very exactly proportional to $T^{3/2}$, but with some additional term near the magnetic transition. The temperature dependence of $\zeta$-Fe and $\zeta$-Co is between $T^2$ and $T^3$. The magnetoresistance of $\zeta$-Mn is large and negative, and peaks at a temperature close to that of the magnetic transition (Fig. 2).

![Fig. 1. — Magnetic susceptibilities of the $\zeta$-phase Mn, Fe and Co compounds; note the ten-fold scale expansion below the axis for $\zeta$-Co. The Curie-Weiss plot for the $\zeta$-Mn data is a best fit to the points above 150 °K.](image)

![Fig. 2. — Magnetization curve of a $\zeta$-Mn sample at 4.2 K.](image)
state (v. b. s.), with the implication that s-d hybridization, and not d-d overlap, is the important interaction. The increased paramagnetism of $\zeta$-Fe with respect to $\zeta$-Co, which corresponds to an additional density of states of $\sim 10 \text{ states/eV per Fe}$, may be attributable to exchange enhancement of the v. b. s. as the magnetic regime is approached. The extraordinarily large g-shift and small saturation moment of $\zeta$-Mn are more difficult to understand.

If these materials were not ordered the resistivity would be large at all temperatures, about 110, 105 and 50 $\mu\Omega\text{cm}$ respectively [5]; at $T = 0$ the resistivity of a perfectly ordered sample vanishes because, to think in virtual bound state terms, the strong scattering at each transitional ion is repeated from unit cell to unit cell and is therefore non-dissipative. The $T^{3/2}$ behaviour of $\zeta$-Mn no doubt represents scattering by spin excitations; the temperature-dependence of the resistivity of $\zeta$-Fe and $\zeta$-Co is too large and too slow to be a phonon term, and it is more likely that it corresponds to the onset of incoherent scattering; at finite temperatures the occupancy of the v. b. s. fluctuates, consequently the scattering phase shift is no longer quite the same in every unit cell, and a finite resistivity appears. This description is essentially equivalent to that of electron-electron scattering in a pure transition metal, but in a situation in which it would be inappropriate to speak of a d band.

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