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Authors
Brock, J.C.F.
Ho, J.C.
Schwartz, G.P.
et al.

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University of California

Ernest O. Lawrence Radiation Laboratory

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LOW-TEMPERATURE HEAT CAPACITIES OF DILUTE SOLUTIONS OF Fe IN Cu.* J. C. F. Brock,† J. C. Ho,‡ G. P. Schwartz, and N. E. Phillips. Inorganic Materials Research Division, Lawrence Radiation Laboratory, and Department of Chemistry, University of California, Berkeley, California 94720.

Most measurements of the heat capacities of dilute magnetic alloys have been interpreted in terms of the indirect (via the conduction electrons) exchange interaction between the magnetic ions. Overhauser[(1)] and Marshall[(2)] have derived, from somewhat different models, similar expressions for the low-temperature heat capacity that predict a concentration-independent term proportional to temperature. Measurements on dilute Fe in Cu by Franck, Manchester and Martin[(3)] above 0.4°K did not show this behavior but it was possible to extrapolate the data to 0°K in a way consistent with the predictions. More recently, experimental and theoretical evidence for the existence of a low-temperature spin-compensated state for isolated localized moments has been obtained,[(4)] and expressions predicting concentration-proportional heat capacities varying (at temperatures well below the Kondo temperature, T_K) as T,[(5,6)] T ln T,[(7)] and $T^{1/2}$ [(8)] have been derived. Separation of the two heat capacity contributions (arising from interactions between magnetic ions and from the thermal break-up of the spin-compensated state) therefore require measurements over a wide range of concentrations. We report here

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† Present address: University of the Witwatersrand, Johannesburg, South Africa.

‡ Present address: Battelle Memorial Institute, Columbus, Ohio.

some preliminary results of new measurements on dilute solutions of Fe in Cu that cover concentration regions in which each contribution is expected to dominate the heat capacity.

The samples were prepared under vacuum in an induction furnace and chill-cast from the melt. Chemical analysis of portions from opposite ends of the samples and microprobe analysis failed to show any evidence of inhomogeneity or precipitation of the Fe. The 4.2-°K resistivity also suggests that the samples are uniform and free of effects of oxidation.

Heat capacity measurements have been made on 0.070, 0.13, 0.18, and 0.27 at% samples between ≈ 0.06°K and 1°K, and on a

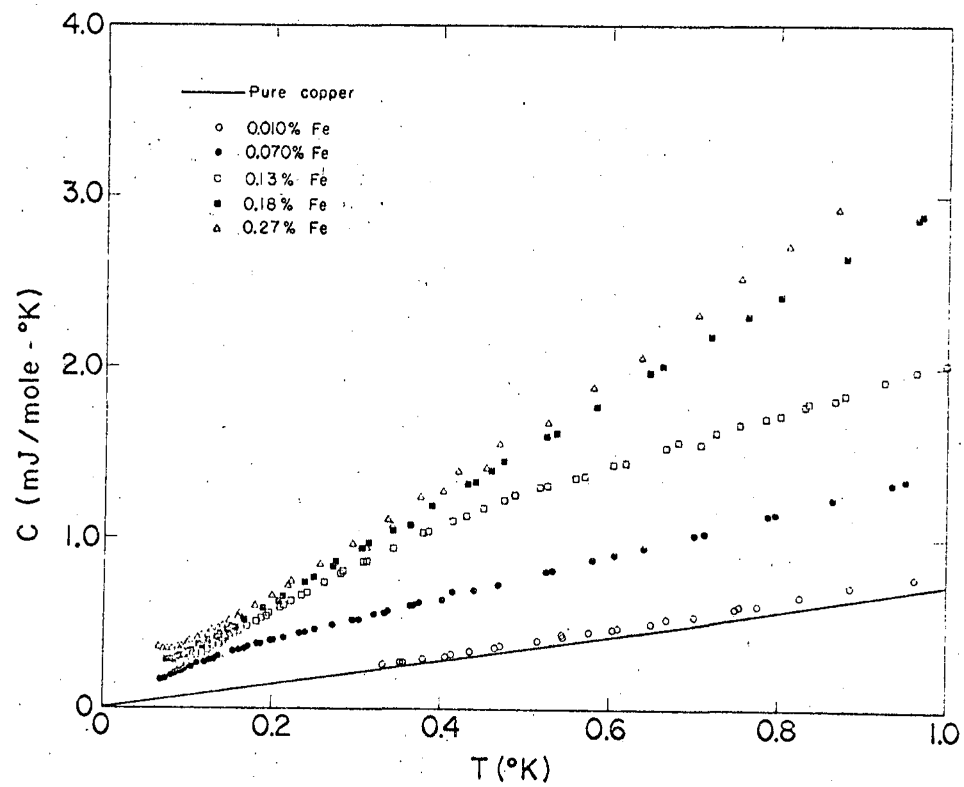

Fig. 1. The low-temperature heat capacities of dilute solutions of Fe in Cu.

0.01 at% sample between 0.3°K and 20°K. The results are shown in Fig. 1. At the lowest temperatures a small T^{-2} term, approximately proportional to concentration, is observed. For the 0.07 to 0.27 at% samples, subtraction of the T^{-2} term leaves a well-defined linear term extending to about 0.11°K for the 0.07 at% sample, and to higher temperatures for the others, in qualitative agreement with the extrapolations made by Franck et al.[3] The linear term however is not concentration independent, but increases by about 40% as the concentration increases from 0.07 to 0.27 at%. This trend is similar to, but larger in magnitude than, that observed by Hill and Pickett[9] at higher temperatures and

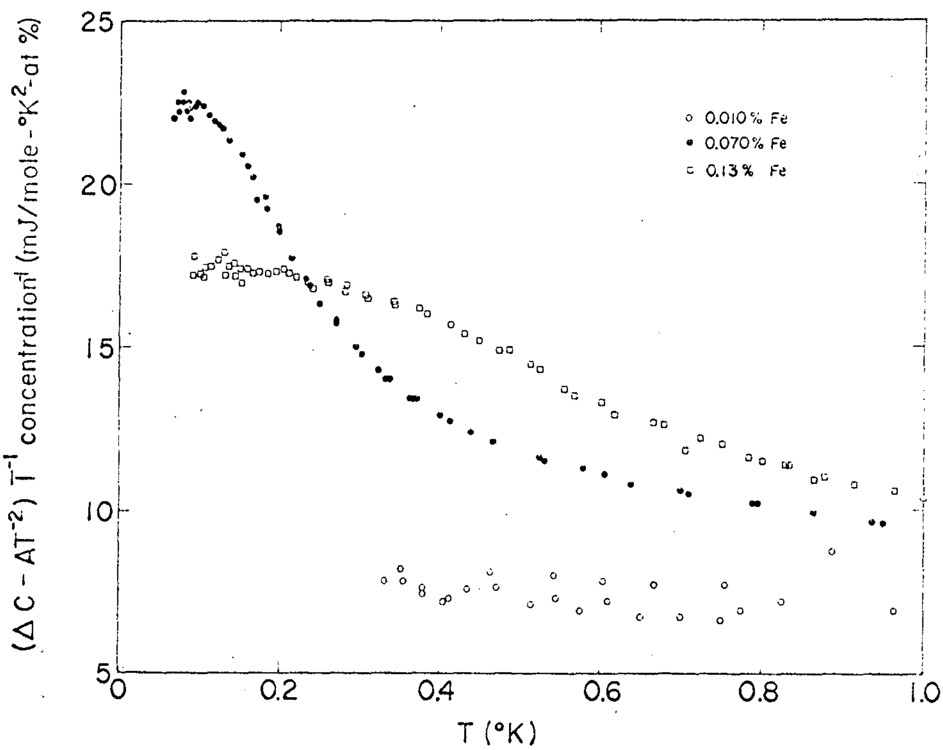

Fig. 2. The excess heat capacities of the dilute solutions (over that of pure copper) divided by T and by concentration.

concentrations. The decrease in the linear term with decreasing concentration may itself be a consequence of the occurrence of the spin compensated state, since the formation of this state, in the more dilute solutions, can be expected to reduce the interactions between the localized moments. At the higher temperatures and lower concentrations the heat capacity in excess of that of pure copper becomes more nearly proportional to concentration (see Fig. 2.), as suggested by the earlier work.[3] The 0.010 at% sample does not appear to show the low-temperature rise in C/T associated with the "concentration-independent" interaction term, and the results for this sample may correspond to the concentration-proportional limit expected for the spin-compensated state. Further experiments are necessary to establish this definitely but it is already clear that in this limit C varies more rapidly than $T^{1/2}$, and may in fact be proportional to T or to T ln T.

Daybell et al.,[10] have recently reported measurements between 0.04 and 1°K on 0.011 and 0.038 at% samples, and interpret the results as showing a $T^{1/2}$ heat capacity for the spin-compensated state throughout this range. The 0.010 at% results reported here are more precise by a factor of 5 than their 0.011 at% results at comparable temperatures and, although they agree in magnitude, show that between 0.3 and 1°K C is varying much more nearly as T. Furthermore, the marked deviations from proportionality to concentration found in this work strongly suggest that the low-temperature upturn in C/T reported by Daybell et al., is associated with interactions between the Fe ions.

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