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B. B. Triplett and Norman E. Phillips

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Low-Temperature Heat Capacity of $Ni_{0.62}Rh_{0.38}$*

B. B. Triplett and Norman E. Phillips

Inorganic Materials Research Division, Lawrence Berkeley Laboratory,
and Department of Chemistry; University of California
Berkeley, California 94720

ABSTRACT

The magnetic field dependence of the anomalous heat capacity of $Ni_{0.62}Rh_{0.38}$ at low temperatures is consistent with superparamagnetic behavior of ferromagnetic clusters, but not with spin fluctuation effects.

Heat capacity anomalies have been observed for a number of transition metal alloys at concentrations near the critical concentration for the appearance of ordered magnetic moments. These anomalies have usually been interpreted as arising from the splitting of the energy levels of superparamagnetic clusters[1,2] by an anisotropy field. For the Ni-Rh system the anomalous heat capacity above 1 K has been shown[3] to have a temperature dependence consistent with theoretical predictions[4] for spin fluctuation effects. However, it has also been suggested[5] that the Ni-Rh data are equally consistent with the heat capacity expected for superparamagnetic clusters. The measurements reported here were undertaken to obtain heat capacity data for a Ni-Rh sample at lower temperatures, and to study the dependence of the anomalous heat capacity on magnetic fields to 38 kOe. The field dependence was expected to provide an additional basis for distinguishing between superparamagnetism and spin fluctuations. A field of 30 kOe should increase the splitting of the energy levels of a cluster to 4 K and reduce the below-1 K heat capacity of clusters considerably, but should have only a small effect on the anomalous heat capacity if it is associated with spin fluctuations.

A $Ni_{0.62}Rh_{0.38}$ sample was prepared as described in Ref. 3, including the 3-day anneal at 1100°C. The heat capacity was measured between 0.05 and 4 K in zero field and between 0.1 and 0.6 K in 3 kOe. The heat capacity was later measured between 0.3 and 25 K in 0, 9, 18, and 38 kOe, but in the meantime the sample had been annealed for an additional 30 days at 1100°C. The anneal increased the heat capacity by between 1 and 3% in the 0.3 to 4 K region. Figure 1 shows the results except that the zero field data taken before the 30 day anneal are omitted above 0.6 K for clarity.

It is evident that the anomalous contribution to the heat capacity is strongly dependent on magnetic field. It has been shown that a field equivalent to the spin fluctuation temperature T_s should be required to suppress the heat capacity contribution associated with spin fluctuations.[4] For $Ni_{0.63}Rh_{0.37}$, $T_s \approx 250$ K has been estimated,[3] and T_s should be similar for $Ni_{0.62}Rh_{0.38}$ since the heat capacities are similar. The observed field dependence is therefore an order of magnitude greater than would be expected on the spin fluctuation model. The behavior shown in Fig. 1 is, however, consistent with that expected for superparamagnetic clusters. (A similar field dependence has been observed in the Fe-V system and interpreted on the basis of clusters.[6]) In zero field the heat capacity of the clusters is expected to be approximately constant over a limited range of temperature, and to drop exponentially to zero below a characteristic temperature that is determined by the anisotropy field.[1,2] In the presence of an external field larger than the anisotropy field the characteristic temperature would be determined by the external field, and, if the free-electron gyromagnetic ratio is assumed for the clusters, should be of the order of 5 K in 38 kOe. Below 10 K, the 38 kOe data are approximately fitted by the expression

$$C = 13.3\,T + 0.067\,T^3 \quad \text{mJ/mole K}, \qquad (1)$$

which suggests that the anomalous heat capacity has been largely shifted to higher temperatures. (The T^3 term and the small positive deviations from Eq. 1 near 2 K probably include some heat capacity of magnetic origin.) We conclude that the field dependence of the anomalous heat capacity is consistent with the superparamagnetism model and not with the spin fluctuation model.

If it is assumed that Eq. 1 is an approximation to the sum of the lattice and electronic heat capacities, the low-temperature low-field magnetic heat capacity can be found by subtracting Eq. 1 from the experimental data. The magnetic heat capacity calculated in this way shows the temperature dependence expected for clusters on the basis of general qualitative considerations[1,2] — a region of approximate independence of temperature, and a sharp drop at lower temperatures. There is also some indication of the expected drop at higher temperatures but this is obscured by the increasing lattice heat capacity.

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Figure Caption

Fig. 1 The heat capacity of $Ni_{0.62}Rh_{0.38}$.

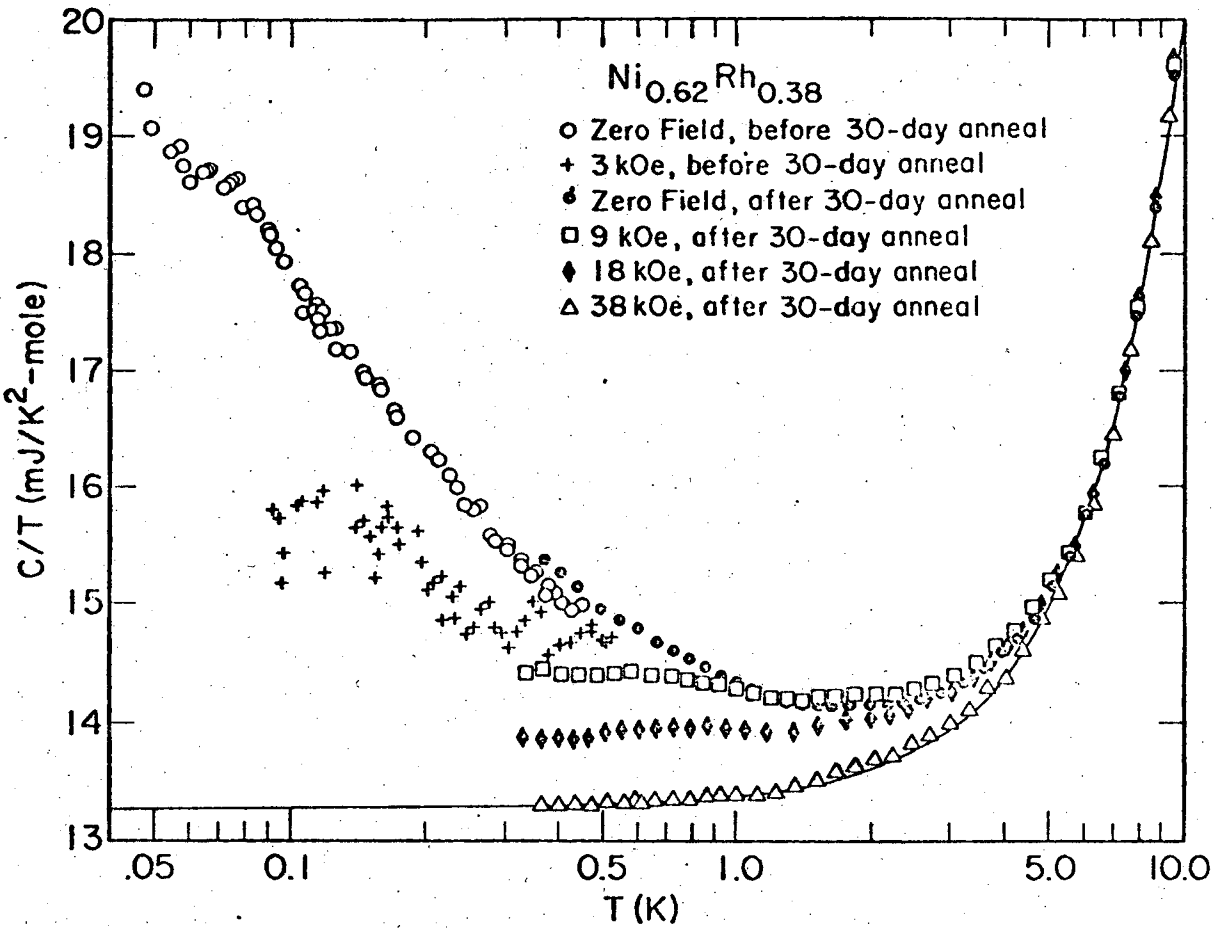

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Fig. 1

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