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Critical Behavior in the Magnetic Ordering of the 95%Ni-2%Mn-2%Al Alloy

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In this work we report a study of the specific heat at normal pressure and at zero magnetic field in the 95%Ni-2%Mn-2%Al alloy near its Curie temperature at $T_C = 463$ K. The specific heat was measured by using high-resolution ac calorimetry technique (under modulated temperature). Adjustments were made by using a $c_P(T)$-function near its Curie critical temperature, T_C. The $c_P(T)$-functionality is a power-serie expansions in α-critical exponent giving by $c_P(T)= \epsilon^{-\alpha}(A_0 + B_0\epsilon^{-\alpha} + A_1\epsilon + B_1\epsilon^{1+\alpha})$, where A_i are the amplitudes and B_i the background contributions. The best values of fitting parameters are: for $T < T_C$ was obtained α=0.34, $A'_0 = 0$, $B'_0 = 1.5$, $A'_1 = 0$, $B'_1 = 1$ with $T_C = 462.1$ K and for $T > T_C$ was obtained α=0.20, $A_0 = 0.48$, $B_0 = -119.8$, $A'_1 = 0$, $B'_1 = 0.27$ for T$_c$ = 464.0 K, showing a slight similarity between the critical exponents when $\epsilon \to 0^-$ and $\epsilon \to 0^+$, respectively. In summary, we report that the critical exponent $\alpha = 027 \pm 007$ is in the range accepted by international literature for other second-order phase transitions for ferromagnetic-paramagnetic ordering (α=0.3). The characteristics of this system near T$_C$ are very similar to that of pure Ni.

Keywords: Specific heat; α-critical exponent; high-resolution ac calorimetry; alumel.

En el presente trabajo reportamos un estudio del calor específico a presión normal y campo magnético cero en la aleación 95%Ni-2%Mn-2%Al cerca de su temperatura de Curie en $T_C = 463$ K, El calor específico se midió usando la técnica de calorimetría ac de alta resolución (bajo temperatura modulada). Los ajustes se hicieron con la función $c_P(T)$ cerca de la temperatura de Curie T_C. La funcionalidad de $c_P(T)$ es una expansión en series en el exponente crítico α de la forma $c_P(T) = \epsilon^{-\alpha}(A_0 + B_0\epsilon^{-\alpha} + A_1\epsilon + B_1\epsilon^{1+\alpha})$, donde A_i son las amplitudes, B_i las contribuciones de base y ϵ es la temperatura reducida, definida como $(T - T_C)/T_C$.
Los valores de los parámetros de ajuste fueron: Para $T < T_C$ se obtuvo $\alpha' = 0.34$, $A'_0 = 0$, $B'_0 = 1.5$, $A'_1 = 0$, $B'_1 = 1$ con $T_c = 462.1$ K y para $T > T_C$ se obtuvo α=0.20, $A_0 = 0.48$, $B_0 = -119.8$, $A'_1 = 0$, $B'_1 = 0.27$ para $T_C = 464.0$ K, mostrando una ligera similitud entre los exponentes críticos cuando $\epsilon \to 0^-$ y $\epsilon \to 0^+$, respectivamente. En resumen, reportamos que el exponente crítico α = 0.27 $\pm$0.07 está dentro del rango aceptado en la literatura internacional para una transición de fases ferromagnética-paramagnetica de segundo orden ($\alpha = 0.3$). Las características de este sistema cerca de T_C son muy similares al sistema níquel puro.

Descriptores: Calor específico; exponente crítico α; calorimetría de alta resolución (ac); alumel.

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1. Introduction

Alumel is an alloy consisting of approximately 95 percent nickel (Ni), 2 percent manganese (Mn), 2 percent aluminum (Al) and 1 percent silicon (Si) that is used for type K-thermocouples. Type K is the most common general purpose thermocouple in the 73 K to 1500 K range. Sensitivity is approximately 41 μV/K. Alumel has an electrical resistivity of approximately 0.294 μ·m.

The Curie temperature of Nickel is T_C = 627 K while the Néel temperature of α-Manganese, which is antiferromagnetic, is T_N = 95 K [1]. However, Alumel, due to its high concentration of Nickel is ferromagnetic whose Curie temperature is about 464 K. One of the striking feature of the ferromagnetic-paramegnetic phase transition, for example in Iron (Fe), whose Tc = 1 044 K, is the critical behavior observed in its thermodynamics properties at zero magnetic field, H = 0, because of the symmetry of a ferromagnet to reverses in the field. There is a characteristic signature of criticality: The specific heat diverge and is infinite at the critical temperature itself [2].

Another common features of the ferromagnetic materials is that the magnetization $m(T)$ below T_C is a decreasing function of T and vanishes at T_C. For T very close to T_C, the power laws behavior

$$m \approx (T_C - T)^\beta$$

is observed, where β is called a critical exponent (β=0.33$\pm$0.03) [3].

The specific heat for a system is specified by its energy levels and is governed by the manner in which the internal energy is distributed among them. The particles of the system can have translational, rotational, vibrational motions, and electronic energy levels, and each type of thermal motions contributes to the specified heat of the system. This contribution gives the total specific heat obtained by various experimental techniques: adiabatic and ac calorimetries.

In this paper, we used the technique of high-resolution ac calorimetry to measure the specific heat at constant pressure of alumel near its magnetic transition around 463 K and use the approximation serial power function for the specific heat as a function of temperature T, c$_P(T)$, to fit the experimental data. The aim is to study the critical behavior of the phase transition near T_C [5].

The outline of the present paper is as follows: In Sec. 2 we present the theoretical details about phase transition and experimental aspects are given in Sec. 3. The results are presented in Sec. 4 and some concluding remarks are given in Sec. 5.

2. Theoretical details

The partition function Z of a material system can be written, in first approximation, by a product of the partition functions for the various contributions of the total energy, *i.e.*,

$$Z = Z_t Z_r Z_v Z_e$$

where Z_t, Z_r, Z_v, and Z_e are the contributions of the translational (t), rotational (r), vibrational (v), and electronic (e) motions, respectively.

However, there is another thermal excitation which occurs over a restricted range of temperatures and contributes to the energetic of the system, thus, to the specific heat too. At temperature much below T_C, the thermal energy is insufficient to cause many excitation, and at higher temperature ($T > T_C$), the levels are equally populated and small change in energy is possible, while as $T \sim T_C$ transitions can occur spontaneously. Therefore, the specific heat is significant high only in the region $T \sim T_C$ and is usually detected as a sharp singularity.

To describe this behavior of a function $f[x]$ as it approaches a nonanalytical ("critical") point x_C, it is introduced the critical exponent λ defined by

$$f[x] \sim (\Delta x)^{\lambda} \quad \text{as} \quad \Delta x = x - x \rightarrow 0^{+} \tag{1}$$

or

$$\lim_{\Delta x \rightarrow 0^{+}} \frac{\ln[f[x]]}{\ln[\Delta x]} = \lambda \tag{2}$$

where $f[x]$ is a nonanalytic function.

The limit (2) does not imply that $f[x]$ is actually proportional to x^{λ}. Fisher [6] proposed three principal cases:

1. *Pure power*

$$f[x] = A(\Delta x)^{\lambda} f_0[x] \quad A = \text{constant};$$

2. *Simple case*

$$f[x] = (\Delta x)^{\lambda} f_0[x]$$

where

$$f_0[x] = f_0 + f_1(\Delta x) + f_2(\Delta x)^2 + \cdots$$

is analytic in the neighbor of $x = x_C$;

3. *Complex case*

$$f[x] = (\Delta x)^{\lambda} f_0[x]$$

but $f_0[x]$ is constant at x = x_C.

For our system, the specific heat exponent α is given by

$$C_p = \begin{cases} \epsilon^{-\alpha}, & \epsilon \rightarrow 0^{+} \\ \epsilon^{-\alpha'}, & \epsilon \rightarrow 0^{-} \end{cases} \tag{3}$$

where

$$\epsilon = \frac{T - T_C}{T_C}$$

is the reduced temperature and fall into class 3, *i.e.*, one can expect to have the following behavior

$$C_p[T] = \epsilon^{-\alpha}(A_0 + B_0\epsilon^{\alpha} + A_1\epsilon + B_1\epsilon^{1+\alpha}) \tag{4}$$

where the amplitudes A and the background B are smooth functions.

3. Experimental details

Alternating Current heating Calorimetry, ACC [4], is a technique for studying phase transitions and generally used to study the thermal behavior (thermal response function of the sample as a function of temperature) of a very small quantity –about 20 mg– of any substance of interest. The substance under study is thermally coupled to a biggest mass called "reservoir" through an inert gas such as helium. The "reservoir" is heated at a constant rate, due to thermal coupling, the sample is heated similarly.

The sample is illuminated periodically with a rich source of infrared light to produce small fluctuations in temperature around the average temperature of the reservoir. Light heating reduces the electrical noise and the thermal inertia of the sample holder, but hardly measures the amount of heat absorbed by the sample. This problem usually is solved by normalizing the data with respect to absolute measurements previously reported in the literature.

The measures are carried out in a frequency regime in which the amplitude of fluctuations in temperature of the sample is inversely proportional to the specific heat. The sample is in tight-vacuum chamber, the entire active space being an almost cylindrical cell 25 mm hight and approximately 20 mm in diameter. On one side of this cell there is a heater that can deliver very precise amounts of heat, and on the other side there is a carbon-type thermistor which measures and controls the temperature of the bath. The electrical resistance of the heater is in the 100 Ω range and the thermistor has a resistance of approximately 1 MΩ at 0°C.

Two 25 μm type-K thermocouples were attached on the rear face of the sample for monitoring its average dc-temperature and its induced temperature oscillations due to the periodic absorption of a small amount of heat on its front face. The sample, with the leads attached was mounted in the chamber, in a horizontal position and close to the thermal bath, to a distance of $\sim$ 0.2 mm. In illuminating the sample chamber care had to be taken to avoid error due to electrical junctions at different temperatures or to other spurious thermoelectrical effects resulting from stray light. Details of the calorimetry set-up are given in Ref. 4. The sample of alumel was obtained from commercial wire (Omega Inc.). The technique of high-resolution ac calorimetry [5,7] was used for measures of specific heat.

4. Results and discussions

Figure 1 shows the experimental data of the thermoelectrical power of the tested sample of alumel, which was taken to verify the critical region of its ferromagnetic-paramagnetic phase transitions (previously reported in Ref. 8).

Ac calorimetry data of the sample of alumel were processed as described in Ref. 8 and in the Fig. 2 exhibits the behavior of the specific heat c_P(T) as a function of absolute temperature T. The measured temperature range was 50 – 700 K for a external field $H = 0$, but only temperature region is shown where the anomaly occurs for a better illustration. The phase transition from ferromagnetic to paramagnetic ordering is of the second-order type or continuous transition, at $T_C = 463$ K, due to the fact that there is no latent heat. The data exhibited a pronounced specific heat peak at $T_C = 463$ K that was significantly sharper than that reported for Niquel [9]. This critical behavior observed near the Curie temperature of Alumel are due to short-range correlations of their magnetic moments. It is a credit to the calorimetry high resolution that such details can be seen near the transition temperature, T_C. It should be stressed that the behavior of cp(T) far from T_C in the low temperature region, is similar to that of Ni [1]. In the same figure it is shown the theoretical fitting using the expression (4).

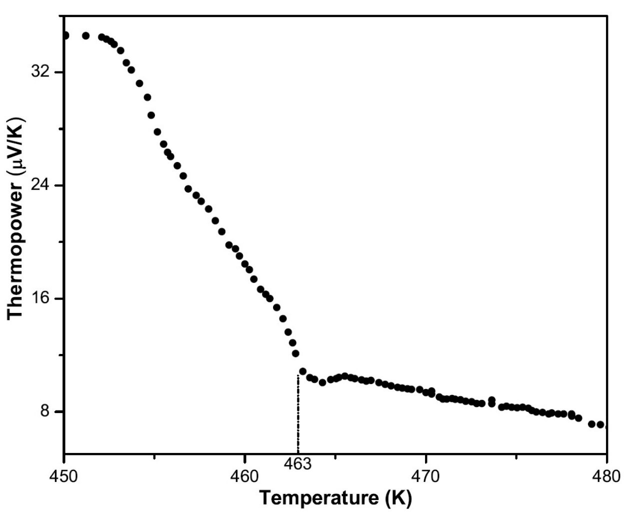

FIGURE 1. Temperature dependence of the thermoelectrical power of alumel close to the ferromagnetic-paramagnetic phase transition.

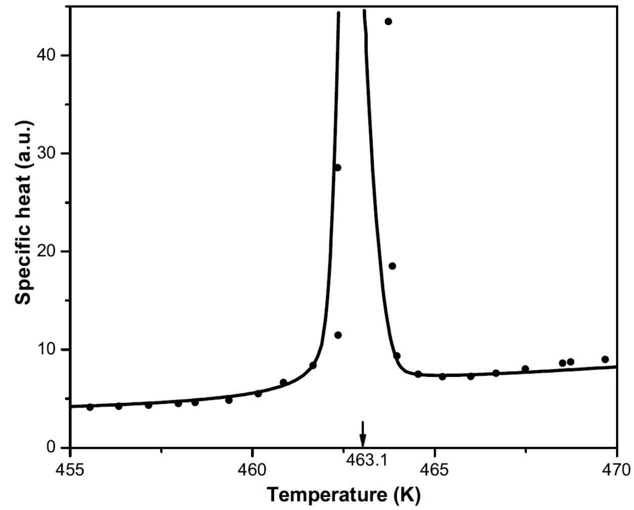

FIGURE 2. Temperature dependence of specific heat $c_P[T]$ under normal pressure as a function of temperature for an sample of alumel, which shows a sharp peak at T_C = 463 K.

The fitting parameters for $T < T_C$ were:

$$A'_0 = 0; \quad B'_0 = 1.5; \quad A'_1 = 0; \quad B'_1 = 1$$

which it gives a fitting value for $\alpha\prime$ = 0.34 and a temperature $T_C = 462.1$ K. The fitting parameters for $T > T_C$ were:

$$A'_0 = 0.48; \quad B'_0 = -191.8; \quad A'_1 = 0; \quad B'_1 = 0.27$$

which it gives fitting value for $\alpha = 0.20$ and a temperature T_C = 464.0 K. and FG-4 samples did not display saturation at H = 27 kOe, this is due to the nanoparticles of magnetite hosted in the fibers, as these present a superparamagnetic behavior and the saturation field is around H = 50 kOe [15].

5. Conclusions

The c_P(T) data for the alumel alloy near the Curie temperature are characteristic of a critical behavior given by a singularity when $T \to T_C$ either from below or above T_C, indicating that correlations are present of all orders.

The fitting parameters $\alpha\prime$ and α are not equal because the range near T_C is very narrow and very sensitive to rounding-off effect of the data.

The average fitting value reported by $\alpha = 0.27 \pm 0.07$ is in the range of values reported by the international literature, for the ferromagnetic-paramagnetic ordering [10].

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