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Magnetic critical scattering in solid $Co_{80}Pd_{20}$

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Abstract

Using small-angle neutron scattering combined with a containerless aerodynamic levitation technique for high temperatures, we have measured the temperature dependence of the correlation length ξ of near-critical magnetic fluctuations in the solid phase of the completely miscible fcc alloy $Co_{80}Pd_{20}$. A fit to our data yields a critical exponent $\nu = 0.76 \pm 0.05$ for the divergence of $\xi(T)$ above the ferromagnetic transition temperature T_c. This value of ν is consistent with the prediction of the three-dimensional Heisenberg model for magnetic critical scattering.

1. Introduction

As evidenced by magnetic susceptibility (Bührer *et al* 2000, Reske *et al* 1995) and electrical resistivity (Lohofer *et al* 2001) measurements, the molten alloy $Co_{80}Pd_{20}$ exhibits an onset of magnetic ordering when undercooled ($T_{\text{liquidus}} = 1606$ K) towards the ferromagnetic transition temperature T_c. The atomic structure of $Co_{80}Pd_{20}$ has been studied by x-ray diffraction in the undercooled liquid phase (Krishnan *et al* 1999) and by extended x-ray absorption fine structure spectroscopy in the solid and undercooled liquid phases (Jacobs and Egry 1999).

Bührer *et al* (2000) confirmed that the magnetic susceptibility $\chi(T)$ does not follow a pure Curie–Weiss behaviour but diverges with a critical exponent γ that can be considered to be a function of temperature (e.g. Fähnle *et al* 1983), and that this temperature dependence $\gamma(T)$ is different for the liquid phase of $Co_{80}Pd_{20}$ and the solid phase. They also observed a difference in transition temperature ($T_{c,\text{liquid}} = 1251$ K versus $T_{c,\text{solid}} = 1272$ K), but the $T \to T_c$ limiting values of γ were consistent in both cases with the value of 1.388 predicted by the classical three-dimensional Heisenberg model for ferromagnets (Stanley 1971, pp 110–115, Collins 1989, p 29).

By using small-angle neutron scattering (SANS) coupled with a high-temperature containerless levitation technique, we have measured the magnetic critical scattering from solid

$Co_{80}Pd_{20}$ in the vicinity of T_c. Here we report for the first time, to the best of our knowledge, experimental results for the temperature dependence of the magnetic (i.e. transverse spin–spin) correlation length $\xi(T)$ of solid $Co_{80}Pd_{20}$ above T_c. For zero applied magnetic field, $\xi(T)$ of a ferromagnetic system should diverge as:

$$\xi(t) = \xi_0 t^{-\nu}, \tag{1.1}$$

where $t = (T - T_c)/T_c$ is the reduced temperature and ν is the critical exponent having a value of 0.707 in the Heisenberg model (Collins 1989, p 29). We obtain a value of $\nu = 0.76 \pm 0.05$ for the solid phase of $Co_{80}Pd_{20}$, consistent with the Heisenberg value (we have yet to perform the experiment on the undercooled liquid phase).

2. Experimental considerations

Our laser-heated aerodynamic levitation technique for neutron diffraction has been described in detail elsewhere (Hennet 2006), including its application to our SANS experiments (Fischer 2007) carried out at the D22 instrument (May and Dewhurst 2006) of the Institut Laue-Langevin in Grenoble, France. For the experiment reported here, the spherical $Co_{80}Pd_{20}$ sample (diameter 2.8 mm and mass 102 mg) was prepared by arc-melting together pieces of ⌀0.5 mm Co wire (99.95 % purity) and ⌀1 mm Pd wire (99.95 % purity). In our SANS set-up, the sample was levitated above a small vanadium nozzle via a vertical flow of purified Ar gas (oxygen limited to 0.1 ppm). A final beam aperture of ⌀4.3 mm, placed a few centimetres upstream of the sample, limited the background scattering. Two adjustable-power 125 W CO_2 lasers of wavelength 10.6 μm heated the sample (one from above, one from below) while a television camera monitored the sample's state and a one-colour pyrometer (wavelength centred on 1.63 μm) recorded the sample's temperature on hard disk every 30 ms.

For small-angle neutron scattering, cobalt's magnetic scattering length of 4.7 fm (Bacon 1975) is in fact larger than its coherent nuclear scattering length of 2.49 fm (Dianoux Lander 2002), permitting a strong magnetic scattering intensity to be observed. Our SANS measurements at D22 used an incident neutron wavelength $\lambda = 4.5$ Å and a detector distance of 4 m matched to a collimation length of 4 m. Counting times were generally only 30 s per SANS image, providing sufficient statistics for a radial average. A very good background subtraction was easily obtained by counting overnight on a cold (i.e. 300 K) levitated sample, since the magnetic SANS intensity from the very large room-temperature magnetic domains occurred at Q values far below our $Q_{min} = 0.02$ Å^{-1}. Such a background subtraction has the advantage of taking into account the sample's incoherent scattering (appreciable for Co), as well as the effect of the sample attenuation on the background intensity. The SANS remaining after background subtraction is essentially only magnetic scattering from the sample. We used the GRASP software package (Dewhurst 2003) for the SANS data treatment.

3. Results and discussion

Figure 1 shows our radially averaged SANS results for the $Co_{80}Pd_{20}$ sample at 1375 K (±10 K). Although the sample was heated to temperatures in excess of 1500 K for almost an hour, we observed no oxidation at its surface afterwards, and the mass was unchanged to within ±0.3 mg. Other than the usual problems involved in maintaining aerodynamic stability of such small levitated samples at high temperature, the main difficulty in our measurements was temperature stability of the sample, as well as the calibration of our one-colour pyrometer (hence the ±10 K temperature uncertainties that we report here). We found that the infrared

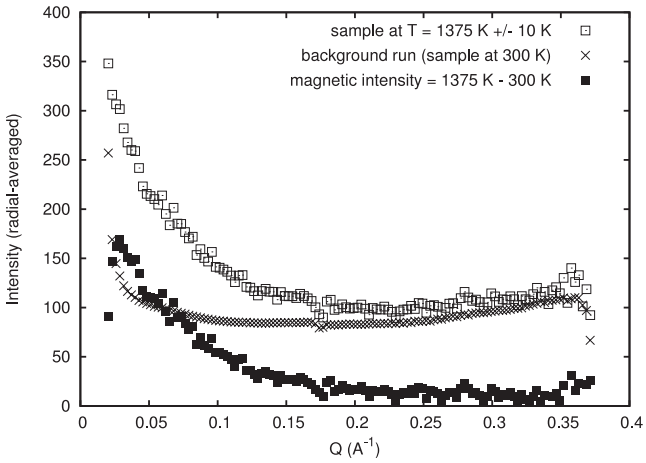

Figure 1. Radially averaged SANS intensity for a levitated solid $Co_{80}Pd_{20}$ sample heated to 1375 K by CO_2 lasers. The same sample levitated at 300 K is used for a background subtraction, since the magnetic intensity from the large ferromagnetically ordered domains at 300 K occurs at Q values well below our measured range ($Q_{\min} = 0.02$ Å^{-1}, $Q_{\max} = 0.37$ Å^{-1}). The counting time for the 1375 K run was only 30 s, for a sample of diameter 2.8 mm and mass 102 mg.

emissivity of $Co_{80}Pd_{20}$ varies as a function of temperature, including a small jump at the solid–liquid transition. We therefore calibrated our one-colour pyrometer afterwards at our laboratory in Orléans against a two-colour pyrometer that is less sensitive to the sample's emissivity: both pyrometers viewed simultaneously a levitated $Co_{80}Pd_{20}$ sample heated over the entire temperature range of our SANS experiment.

As discussed by Shirane *et al* (1984), the scattering law $S(Q, \omega)$ for magnetic scattering from itinerant (i.e. metallic) ferromagnets follows, over a wide temperature range both near and far from T_c, a rather simple product of Lorentzians:

$$S(Q, \omega) \propto \frac{1}{\kappa^2 + Q^2} \frac{\Gamma}{\Gamma^2 + \omega^2}, \tag{3.1}$$

where $1/\kappa = \xi$ is the transverse spin–spin correlation length of magnetic fluctuations. The line width Γ satisfies the relation $\Gamma = \Lambda(T)\ Q^2$ within the so-called hydrodynamic regime ($Q\xi < 4$) that concerns our SANS measurements, where $\Lambda(T)$ is a temperature-dependent spin diffusion constant. When the incident neutron energy exceeds sufficiently the line width Γ, as we have estimated for our case, the SANS measurement performs an adequate integration of the scattering law over ω, resulting in (Shirane *et al* 1984)

$$S(Q) = S_0 \frac{T}{T_c} \frac{1}{\kappa^2 + Q^2}, \tag{3.2}$$

where S_0 is a temperature-independent constant.

To extract ξ from our SANS data, therefore, it is not necessary to normalize $S(Q)$ on an absolute scale but simply to fit the expression for the scattered intensity:

$$I(Q) = I_0 \frac{1}{\kappa^2 + Q^2}, \tag{3.3}$$

which is most conveniently performed by using an Ornstein–Zernicke (OZ) plot as shown in figure 2, where the straight-line fit intersects the abscissa at $-\kappa^2$. We found that the prefactor I_0 obtained from our OZ fits obeyed a linear dependence in T to within experimental uncertainties

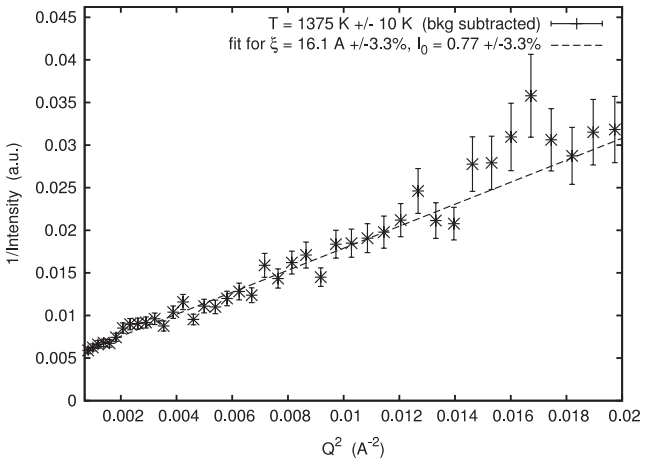

Figure 2. Ornstein–Zernicke plot for the data of figure 1, used to extract a magnetic (transverse spin–spin) correlation length of $\xi = 16.1$ Å $\pm 3.3\%$. Note that the range of this fit covers $Q\xi < 2.3$, which satisfies the standard criterion of $Q\xi < 4$ for the hydrodynamic regime where the Ornstein–Zernicke Lorentzian function is valid for critical scattering.

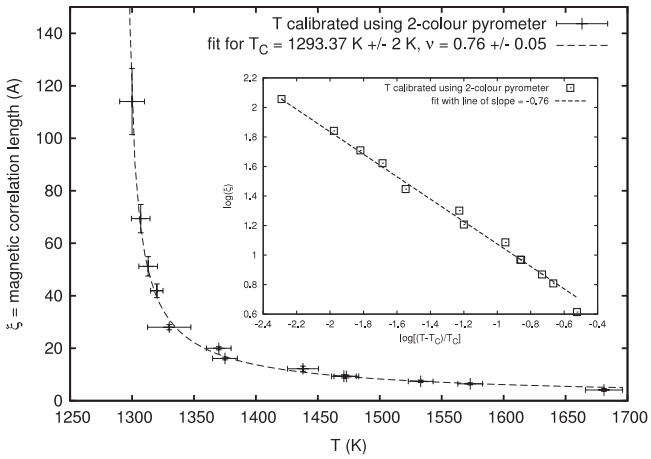

Figure 3. Divergence of the magnetic (transverse spin–spin) correlation length ξ in solid $Co_{80}Pd_{20}$ above the ferromagnetic transition temperature T_c as measured by SANS, including a least-squares fit to $\xi(t) = \xi_0 t^{-\nu}$, where $t = (T - T_c/T_c)$ is the reduced temperature. The data were acquired from left to right, i.e. the sample was continually heated throughout the experiment, except for occasional temperature fluctuations of ∼20 K with time constant ∼30 s, due to small instabilities in the laser heating. The sample was therefore in the solid phase except for the highest temperature data point at 1681 K ($T_{\text{liquidus}} = 1606$ K). The log–log plot of the inset confirms that the exponential divergence of $\xi(t)$ holds true across almost two orders of magnitude in reduced temperature, to within 0.5% of our fitted value of T_c, with the possible exception of the single liquid-phase data point at the far right.

(the exact amount of sample visible to the neutron beam is not well-known and varies somewhat due to thermal expansion effects).

Figure 3 shows our results for the temperature dependence of the magnetic correlation length $\xi(T)$ in the solid phase of $Co_{80}Pd_{20}$. A least-squares fit to equation (1.1) gives $T_c = 1293.37 \pm 2$ K, with a critical exponent $\nu = 0.76 \pm 0.05$, and $\xi_0 = 2.05 \pm 0.25$ Å.

Our value for ν is therefore consistent, within experimental uncertainty, with the Heisenberg value of 0.707. Our value for T_c is only about 21 K greater than that found by Bührer *et al* (2000) for solid $Co_{80}Pd_{20}$, and can be attributed to residual systematic error in our sample temperature measurement.

4. Summary and conclusions

Using an aerodynamic levitation technique with laser heating, we obtained high-quality SANS data for magnetic critical scattering in solid $Co_{80}Pd_{20}$, in spite of the very small sample size ($\varnothing$2.8 mm) and brief counting time (30 s) per data point. Our magnetic SANS intensity obeys well the expected Ornstein–Zernicke form for critical scattering, leading to a robust determination of the magnetic (transverse spin–spin) correlation length ξ. Although the data analysis is somewhat limited by remaining uncertainties in the sample temperature, we observe a clear divergence of $\xi(T)$ near the ferromagnetic transition temperature T_c, and our obtained critical exponent $\nu = 0.76 \pm 0.05$ agrees with the three-dimensional Heisenberg model for ferromagnets. An experiment to determine ν for the undercooled liquid phase of $Co_{80}Pd_{20}$ is planned for the near future, and will include improvements in measurement of the sample temperature.

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