The spin-glass state in the random-exchange $Pd_xCo_{50}Al_{50-x}$ system

2000 J. Phys.: Condens. Matter 12 1819

(http://iopscience.iop.org/0953-8984/12/8/324)

J. Phys.: Condens. Matter **12** (2000) 1819–1825. Printed in the UK PII: S0953-8984(00)06929-0

The spin-glass state in the random-exchange $Pd_xCo_{50}Al_{50-x}$ system

K Eftimova† and J J McGuire
Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada
E-mail: `ktzalikian@hotmail.com`

Received 13 August 1999, in final form 20 December 1999

Abstract. The nonlinear susceptibilities of the random-exchange $Pd_5Co_{50}Al_{45}$ and $Pd_8Co_{50}Al_{42}$ alloys are derived from dc magnetic measurements. They are investigated in the critical region and spin-glass exponents characterizing these materials are obtained. A progressive development of a canonical spin-glass phase is traced from $Pd_5Co_{50}Al_{45}$ to $Pd_8Co_{50}Al_{42}$. The conventional spin-glass phase in $Pd_8Co_{50}Al_{42}$ is characterized with the exponents $\phi = 3.25$ and $\delta = 6.4$ and a phase transition temperature $T_g = 25.3$ K.

1. Introduction

The $Co_{50}Al_{50}$ alloy is known to be strongly paramagnetic [1]. When doped with Pd for Al up to 9 at.% [2], the $Pd_xCo_{50}Al_{50-x}$ system develops a clearly observable spin-glass-like state. In a general magnetic characterization of these alloys [2] the freezing of the magnetic moments below a characteristic temperature, T_f, showed up as a peak in the temperature dependence of the zero-field-cooled magnetic susceptibility, $\chi_{ZFC}(T, H)$. The position of this peak gave $T_f \approx 13.5$ K and 27 K for $Pd_5Co_{50}Al_{45}$ and $Pd_8Co_{50}Al_{42}$ respectively. It was also observed that their χ_{ZFC} and field-cooled magnetic susceptibilities, χ_{FC}, break away from one another.

The $Pd_xCo_{50}Al_{50-x}$ materials ($x = 5$, 8) have small positive Curie–Weiss temperatures. Compared to the $Co_{50+x}Al_{50-x}$ system, their spin-glass-like state is retarded in its development with doping [2, 3].

The question of whether this state in the $Pd_xCo_{50}Al_{50-x}$ alloys ($x = 5$, 8) is a true spin-glass (SG) phase and whether their freezing temperatures, T_f, correspond to a phase transition from the SG phase to the paramagnetic phase upon heating remained open in [2].

This work deals with the nature of the SG freezing phenomenon [4, 5] in the metallic $Pd_xCo_{50}Al_{50-x}$ system ($x = 5$, 8). The adopted approach to investigate this problem is the static scaling of the nonlinear susceptibility, $\chi_{nl}(T, H)$ [5–7]. A multiple field-cooled dc magnetization measurement procedure [6, 7] is used as the experimental method.

2. Theoretical background

It is known [6–8] that the total nonlinear susceptibility of an SG can be expressed as:

$$\chi_{nl}(T, H) = \chi_0(T) - M(T, H)/H. \qquad (1)$$

---

† Addressee for correspondence: Dr K Eftimova, 34 Holmes Avenue, Hamilton, Ontario, Canada L8S 2K6.

0953-8984/00/081819+07$30.00

In equation (1) $\chi_0(T)$ is the linear susceptibility, $M(T, H)$ is the magnetization of the SG and T and H are the temperature and the applied dc magnetic field.

As the total magnetic susceptibility of the SG and its linear term are non-divergent, the relevant physical quantity to identify a phase transition at the temperature T_g is the nonlinear susceptibility (or any of its terms $\chi_i(T)$ $(i = 1, 3, 5, \ldots)$) [8]. The role of the first nonlinear term $\chi_1(T)$ is dominant in low magnetic fields.

It is necessary to separate the linear and nonlinear components of the data. $\chi_0(T)$ and $\chi_1(T)$ can be obtained simultaneously for each temperature point of the measurement, T_i, by fitting the low-field total magnetic susceptibility isotherms, $\chi(T_i = \text{const}, H)$, to the expression $\chi_0(T_i) - \chi_1(T_i)H^2$ [6]. The increase of $\chi_1(T)$ when cooling from paramagnetic temperatures to the critical point should be three orders of magnitude for a typical SG [6, 9]. $\chi_0(T)$ can also be obtained by extrapolating the low-field magnetic susceptibility isotherms to zero field, $\chi(T_i = \text{const}, H \to 0)$ [10].

To describe the behaviour of $\chi_{nl}(T, H)$ in the critical region and to try to determine some SG exponents, the nonlinear susceptibility isotherms, $\chi_{nl}(T_i = \text{const}, H)$, are scaled according to the relation [6, 11]:

$$\chi_{nl}(T, H) \propto H^{2/\delta} f[\tau/H^{2/\phi}] \tag{2}$$

where

$$f(x) = \text{const} \qquad \text{with } x \to 0$$
$$f(x) = x^{-\gamma} \qquad \text{with } x \to \infty.$$

In equation (2) the reduced temperature, defined as $T/T_g - 1$, is denoted by τ, and δ, ϕ and γ are critical exponents of the SG transition.

The critical exponent δ can therefore be obtained from the log–log plot of $\chi_{nl}(T = T_g, H)$ against H. This value of δ is then used in equation (2) to scale the nonlinear susceptibility isotherms for $T > T_f$. The free parameters in equation (2), T_g and ϕ, are varied to obtain the best scaling of the data.

The susceptibility exponent γ can be estimated in two ways in this calculation procedure. The critical exponents γ, δ and ϕ are related through the hyperscaling relation [11, 12]

$$\gamma = \phi(1 - 1/\delta). \tag{3}$$

γ can also be obtained from the scaled data when $x \to \infty$ since, according to equation (2), the value of the asymptotic slope is $-2\gamma/\phi$ [6]. Both ways of estimation should result in similar values of γ. This is one criterion for determining if the investigated material is an SG [6].

3. Experiment

The magnetic measurements were made with a commercial Quantum Design (San Diego) SQUID magnetometer in a temperature interval from 4.2 K to 100 K and in dc magnetic fields up to 5.6 MA m^{-1} (7 T).

Multiple field-cooled dc magnetization measurements against T are used to obtain the points in the field-temperature space as in [6, 7].

The samples were initially cooled down in zero field from room temperature to a temperature not less than 3 T_f. Then a magnetic field in the range of 80 A m^{-1} to 5.6 MA m^{-1} (1–7000 Oe) is switched on, and the $M_{FC}(T, H_i = \text{const})$–$T$ dependence is measured at a fixed applied field down to a temperature below T_g. The sample is then warmed back in the same field to $T > 3\ T_g$ before the next field is switched on and the next M_{FC}–T data are recorded.

4. Results and discussion

4.1. Static scaling for $Pd_5Co_{50}Al_{45}$

In figure 1 are shown the general magnetic characteristics of $Pd_5Co_{50}Al_{45}$. Figure 1(a) displays the temperature dependence of the total magnetic susceptibility $\chi_{FC}(T, H_i = \text{const})$ measured in different constant applied fields, and figure 1(b) presents the magnetic field dependence of $\chi_{FC}(T_i = \text{const}, H)$ for the temperatures of $T = 15$, 16.5, 19.5, 30 and 70 K.

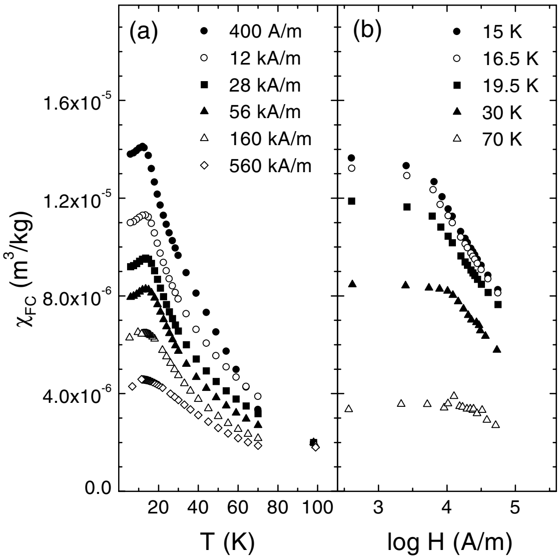

Figure 1. (a) Temperature dependences of the total dc field-cooled magnetic susceptibilities, $\chi_{FC}(T, H_i = \text{const})$ of $Pd_5Co_{50}Al_{45}$ measured using fields in the range of 400 A m^{-1} to 560 kA m^{-1}. (b) The log H dependences of $\chi_{FC}(T_i = \text{const}, H)$ for $Pd_5Co_{50}Al_{45}$ for the indicated temperatures.

While figure 1(a) shows that the role of the nonlinear susceptibility $\chi_{nl}(T, H)$ increases in high dc fields, figure 1(b) demonstrates that the role of $\chi_{nl}(T, H)$ decreases at $T \gg T_f$. The form and the magnitude of the peak in figure 1(a) are changed due to the influence of the nonlinear part of the susceptibility.

The linear term, $\chi_0(T_i)$, for each measured temperature, T_i, is determined by extrapolating the total susceptibility to a zero field value, $\chi_{FC}(T_i = \text{const}, H \to 0)$. Then the magnetic field dependence of the nonlinear susceptibility isotherm $\chi_{nl}(T_i = \text{const}, H)$ is calculated according to equation (1).

The SG exponent δ is obtained from the slope of the log–log plot of $\chi_{nl}(T_i = \text{const}, H)$ against H at $T = 13.5$ K which is the nearest temperature to the temperature of the maximum in the $\chi_{FC}(T, H_i = \text{const})$–$T$ dependence measured in low fields. The linear dependence of $\log \chi_{nl}(T_i = \text{const}, H)$–$\log H$ at 13.5 K, shown in the inset of figure 2, is valid for fields from 64 to 560 kA m^{-1} and gives the value of 7.35 for δ.

The value of $\delta = 7.35$ is substituted into equation (2). The parameters T_g and ϕ are varied, and the parameter δ is further adjusted, to obtain the best scaling shown in figure 2. The values of the parameters obtained from figure 2 are $T_g = 12.7 \pm 0.2$ K, $\phi = 1.6 \pm 0.2$ and $\delta = 7.1 \pm 0.4$.

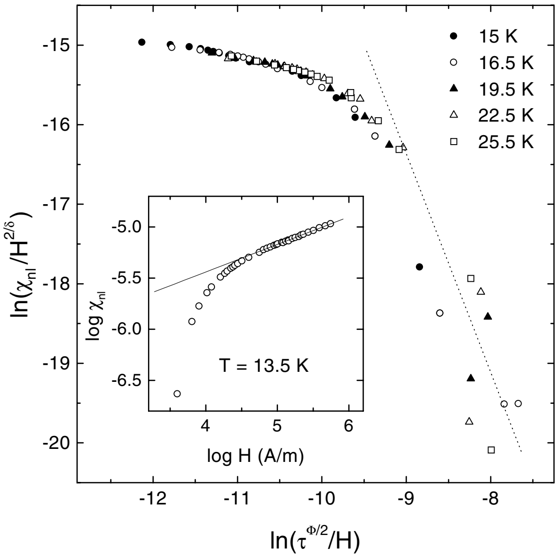

Figure 2. Scaling plot of the nonlinear susceptibility of $Pd_5Co_{50}Al_{45}$ according to equation (2) made for $\chi_{nl}(T_i = \text{const}, H)$ within the temperature interval $1.1\ T_g < T \leqslant 2\ T_g$. The inset shows the $\log \chi_{nl}$–$\log H$ plot for 13.5 K from which the value of δ is determined.

The value of the susceptibility exponent, γ, obtained by equation (3) is 1.38 while the value of γ obtained by the slope of the scaled data when $x \to \infty$ is 1.76. The difference between these two values of γ is 27%.

The calculation of the first nonlinear term of the susceptibility, $\chi_1(T)$, showed the increase of $\chi_1(T)$ from 45 K to 15 K to be a factor of about 10, which is small for typical SGs [6, 9].

Although the scaling gives finite values of the free parameters, the data do not tend to a limiting value when $x \to 0$. Also, the low unphysical values of ϕ and γ and the poor quality of the scaling displayed in figure 2 do not support the conclusion that an SG phase transition occurs in this material.

4.2. Static scaling for $Pd_8Co_{50}Al_{42}$

Figure 3 presents the general magnetic behaviour of the $Pd_8Co_{50}Al_{42}$ alloy.

Figure 3(a) shows the temperature dependences of $\chi_{FC}(T, H_i = \text{const})$ of $Pd_8Co_{50}Al_{42}$ measured in fixed fields in the range of 310 A m^{-1}–350 kA m^{-1}. When the experiment is performed in moderate fields, the magnitude of the peak at $T_f \sim 27$ K is considerably diminished due to the contribution of the nonlinear susceptibility.

Figure 3(b) shows the magnetic field dependences of $\chi_{FC}(T_i = \text{const}, H)$ of $Pd_8Co_{50}Al_{42}$ for some temperatures above T_f. As in figure 1(b), the decreasing role of the nonlinear susceptibility at temperatures $T \gg T_f$ is clearly observed.

The temperature dependences of the linear susceptibility $\chi_0(T)$ and the first nonlinear term $\chi_1(T)$ are obtained by fitting the low field total susceptibility isotherms to the expression $\chi_0 - \chi_1 H^2$. For comparison, the $\chi_0(T)$ data were also obtained by the zero field extrapolation of the total susceptibility isotherms. The results show non-divergent temperature dependence of the linear susceptibility, and the values of χ_0 obtained by the expression $\chi_0 - \chi_1 H^2$ and by the zero field extrapolation do not differ by more than 5% for each temperature point.

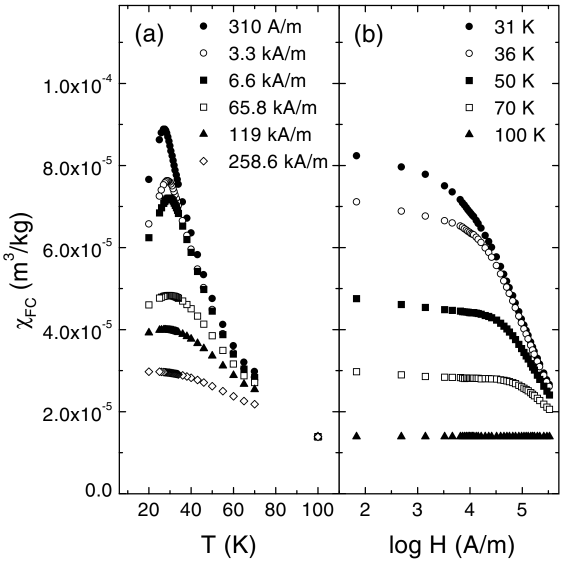

Figure 3. (a) Temperature dependences of the total dc field-cooled magnetic susceptibilities, $\chi_{FC}(T, H_i = \text{const})$ of $Pd_8Co_{50}Al_{42}$ measured using fields in the range of 310 A m^{-1} to 350 kA m^{-1}. (b) The log H dependences of $\chi_{FC}(T_i = \text{const}, H)$ for $Pd_8Co_{50}Al_{42}$ for the indicated temperatures.

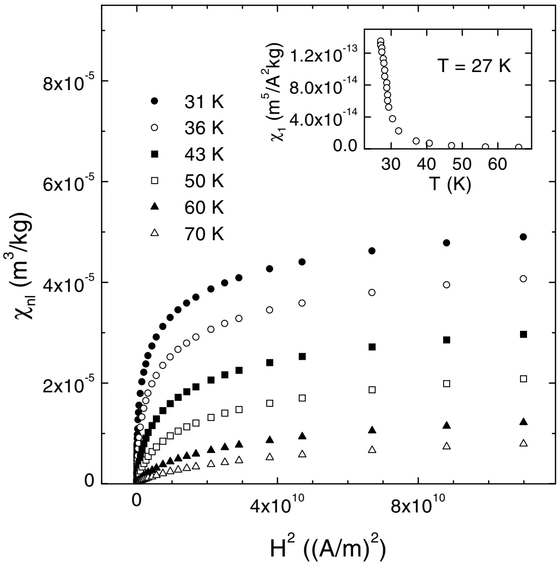

Figure 4. The derived nonlinear susceptibility isotherms of $Ho_8Co_{50}Al_{42}$, $\chi_{nl}(T_i = \text{const}, H)$, against H^2 for the indicated temperatures. The inset shows the χ_1 coefficient against T.

The temperature dependence of the calculated $\chi_1(T)$ term shows divergence at $T \approx 27$ K (see the inset of figure 4). It decreases in magnitude on heating up to 100 K by a factor of about 500.

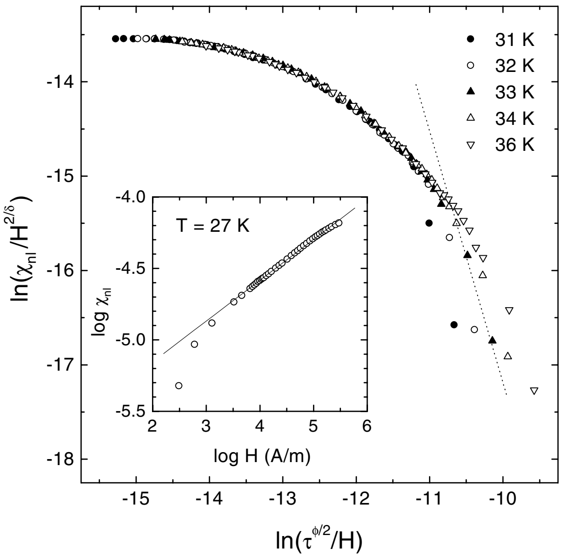

Figure 5. The $\ln(\chi_{nl}/H^{2/\delta})$–$\ln(\tau^{\phi/2}/H)$ scaling of the $\chi_{nl}(T_i = \text{const}, H)$ data for some temperatures in the interval $1.1\ T_g < T < 1.5\ T_g$. The parameters used for this best data collapse are $T_g = 25.3$ K and $\phi = 3.25$. The inset shows the $\log \chi_{nl}$–$\log H$ dependence for $T = 27$ K, from which the value of $\delta = 6.8$ is determined.

Figure 4 displays the dependence of the derived nonlinear susceptibility, $\chi_{nl}(T, H)$, on H^2 for the temperatures of 31, 36, 43, 50, 60 and 70 K. The figure shows a low-field region of linear dependence of the nonlinear susceptibility on H^2 where the $\chi_1(T)$ term is predominant and a notable decrease in the magnitude of the nonlinear susceptibility with temperature increase.

The log–log plot of χ_{nl} against H for $T = 27$ K is shown in the inset of figure 5. This is the temperature of the maximum in the χ_{FC}–T plots measured in low fields. From the linear regression in the field region of 6.6 to 350 kA m^{-1}, the value of the exponent δ is found to be $\delta = 6.8 \pm 0.1$. The deviation from a linear dependence for fields lower than 6.6 kA m^{-1} can be attributed partly to a larger experimental error in the values of small fields.

The value $\delta = 6.8$ is used in equation (2) to scale the $\chi_{nl}(T, H)$ data for the field region of the measurement (310 A m^{-1}–350 kA m^{-1}) and for temperatures $T > T_f$. Figure 5 presents the best data collapse in the $\ln(\chi_{nl}/H^{2/\delta})$–$\ln(\tau^{\phi/2}/H)$ scale for some temperatures in the range $1.1\ T_g < T < 1.5\ T_g$. The parameters used for this best collapse are $T_g = 25.3 \pm 0.1$ and $\phi = 3.25 \pm 0.15$, and the adjustment of δ resulted in $\delta = 6.4 \pm 0.3$.

The calculated values of the first nonlinear term $\chi_1(T)$ were tested for the obtained value of the transition temperature T_g. The $\log \chi_1(T)$–$\log(T/T_g - 1)$ dependence resulted in a good-fit straight line for $T_g = 25.3$ K.

With these values of ϕ and δ equation (3) gives the susceptibility exponent $\gamma = 2.74$, and the SG ordering parameter $\beta = \phi/\delta \approx 0.51$ [6, 11]. The calculation of related SG exponents [8] gives the specific heat exponent $\alpha = 2 - 2\beta - \gamma = -1.76$ and the exponent of the correlation length $\nu = (2 - \alpha)/d = 1.25$ where d is the dimensionality. These values for $Pd_8Co_{50}Al_{42}$ are on the low end of the range of values of the exponents of other SG materials [6–8]. The negative value of α, which is in accordance with results for other SGs, indicates that there is no divergence in the specific heat since it scales as $\tau^{-\alpha}$ [13].

The scaling of the data presented in figure 5 was tested at the limiting points. It is seen from figure 5 that in the limit of $x \to 0$ the data tend to an asymptotic constant value.

The asymptotic slope of the scaled data in figure 5 when $x \to \infty$ is 1.71 with variation of this value less than 1% for each isotherm. This slope results in the value of $\gamma = 2.89$, which is in very good agreement with the value of γ obtained using equation (3).

Thus, from the point of view of the scaling theory it seems possible to conclude that a paramagnetic → SG phase transition occurs in $Pd_8Co_{50}Al_{42}$ at the temperature of $T_g = 25.3$ K. This SG alloy is characterized with the exponents $\phi = 3.25$ and $\delta = 6.4$.

5. Conclusions

The static scaling analysis performed in this work leads to the conclusion that the magnetic state below T_f of $Pd_5Co_{50}Al_{45}$ does not correspond to an SG phase. In support of this conclusion are the nonphysical values of the exponents obtained from the scaling analysis and the small increase (~tenfold) of the $\chi_1(T)$ term upon cooling from paramagnetic temperatures.

For the $Pd_8Co_{50}Al_{42}$ alloy the increase of $\chi_1(T)$ as the critical temperature is approached from above is a factor of about 500, which is typical for a paramagnetic → SG transition. The obtained values of the SG exponents are also in the range of reported values [6–8, 10, 11] for other SG materials, although they are on the low side indicating that 8 at.% of Pd is near the onset of SG behaviour for the $Pd_xCo_{50}Al_{50-x}$ system.

While doping with 5 at.% of the 4f element Ho resulted in the formation of an SG phase in $Ho_5Co_{50}Al_{45}$ [14], doping with 5 at.% of Pd leads only to a prelude of an SG phase in $Pd_xCo_{50}Al_{50-x}$ with true SG behaviour emerging near 8 at.%.

In [2] a model of the substitution of the Pd atoms in the CsCl crystal lattice was developed. Obviously at 8 at.% of Pd the quantity of the displaced Co atoms leads to SG interactions and qualitative formation of an SG phase.

Acknowledgments

The research was supported by NSERC Canada. Wihuri Physical Laboratory of the University of Turku is acknowledged for their kindness in providing this investigation with $Pd_xCo_{50}Al_{50-x}$ alloys.

References

[1] Sellmyer D, Caskey G and Franz J 1972 *J. Phys. Chem. Solids* **33** 561

[2] Eftimova K, Laiho R, Lisunov K G and Lahderanta E 1996 *J. Magn. Magn. Mater.* **154** 193

[3] Lahderanta E, Eftimova K, Laiho R, Al Kanani H and Booth J G 1994 *J. Magn. Magn. Mater.* **130** 23

[4] Chalupa J 1977 *Solid State Commun.* **24** 429

[5] Suzuki M 1977 *Prog. Theor. Phys.* **58** 1151

[6] Martinez B, Labarta A, Rodriguez-Sola R and Obradors X 1994 *Phys. Rev.* B **50** 15 779

[7] Gingras M J P, Stager C V, Raju N P, Gaulin B D and Greedan J E 1997 *Phys. Rev. Lett.* **78** (5) 947

[8] Fischer K H and Hertz J A 1991 *Spin Glasses* (Cambridge:Cambridge University Press)
Binder K and Young A P 1986 *Rev. Mod. Phys.* **58** 801

[9] Omari R, Prejean J J and Souletie J 1983 *J. Physique* **44** 1069

[10] Gunnarsson K, Svedlindh P, Nordblad P and Lundgren L 1991 *Phys. Rev.* B **43** 8199
Novak M A, Symko O G and Zheng D J 1986 *Phys. Rev.* B **33** 343

[11] Barbara B, Malozemoff A P and Imry Y 1981 *Phys. Rev. Lett.* **47** 1852
Malozemoff A P and Barbara B 1985 *J. Appl. Phys.* **57** 3410

[12] Gavrin A, Childress J R, Chein C L, Martinez B and Salamon M B 1990 *Phys. Rev. Lett.* **64** 2438

[13] Brodale G E, Fisher R A, Fogle W E, Phillips J O and van Curen J 1983 *J. Magn. Magn. Mater.* **31–34** 1331

[14] Eftimova K, Lahderanta E and Laiho R 1999 *J. Phys.: Condens. Matter* **11** 6935