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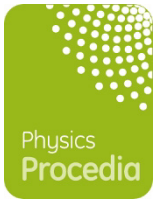

Magnetic properties of the $RAuBi_2$ (R = Ce, Pr, Nd, Gd, Sm) series of intermetallic compounds

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Abstract

In this work we report the physical properties of the series of intermetallic compounds $RAuBi_2$ (R = Ce, Pr, Nd, Gd, Sm) studied by means of x-ray powder diffraction, magnetic susceptibility, heat capacity and electrical resistivity measurements. These compounds were grown using the metallic Bi self-flux technique and they crystallize in a tetragonal $ZrCuSi_2$ (P4/nmm) structure. Our $RAuBi_2$ single crystals show an antiferromagnetic ordering $T_N \geq 2.3$ K for all measured compounds. The values of T_N show a dramatic breakdown of the De Gennes factor along this series.

Keywords: Intermetallic compounds, crystalline electrical field (CEF), magnetic interaction

1 Introduction

Rare-earth based intermetallic compounds are subject of great interest to the scientific community due to a wide variety of interesting physical properties, such as: crystalline electrical field (CEF) effects, Ruderman-Kittel-Kasuya-Yosida (RKKY) magnetic interactions, Kondo effect, heavy fermion (HF) behavior and unconventional superconductivity. [1, 2, 3, 4, 5] Systematic studies along series of rare-earth based intermetallic compounds usually offer the opportunity to explore the interplay among all these effects. Well known examples of emblematic studies in series of structurally related compounds have been performed for the intermetallic $RMIn_5$ (115, M = Rh, Ir, Co) (and relative compounds) and RM_2T_2 (122 system; M = e.g. Fe, Cu, Ni, Rh, Ru; T = e.g. Si, Ge, As). [6, 7, 8, 9, 10, 11, 12, 13]

Interestingly, several Ce-based heavy fermions superconductors $CeMIn_5$ (M = Co, Rh, Ir), Ce_2MIn_8 (M = Co, Rh, Pd), $CePt_2In_7$ and $CeCu_2Si_2$ [8, 14, 15, 16] form in tetragonal crystal structure and therefore it is generally believed that such low symmetry structures may favor the occurrence of such remarkable phenomena. Therefore, the study of structurally related series

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of compounds is motivated by the possibility of obtaining new heavy fermions superconductors. Furthermore, is a great opportunity to explore the evolution of the properties along its members.

In this context, we have studied a new family of intermetallic compounds $RAuBi_2$ (R = Ce, Pr, Nd, Sm, Gd) , which also crystallize in a tetragonal structure with a $ZrCuSi_2$-type structure (space group P4/nmm) in order to explore the evolution of the magnetic properties along this series. In a recent publication, [17] we explore the details of the magnetic properties of our single crystals of nearly stoichiometric $CeAuBi_2$. This compounds orders antiferromagnetically at $T_N = 12$ K with a magnetic anisotropy consistent with an Ising antiferromagnet and the field dependent magnetization data at low temperatures reveal the existence of a spin-flop transition when the magnetic field is applied along the c-axis ($H_c \approx 7.5$ T and T = 5 K). In this paper, we extent these results to other rare-earths through detailed measurements on single crystals $RAuBi_2$ grown from Bi-flux by means of x-ray powder diffraction, magnetic susceptibility, electrical resistivity, and heat capacity measurements. The behavior of antiferromagnetic ordering temperature T_N shows a dramatic breakdown of the De Gennes factor along this series.

2 Experiment

$RAuBi_2$ (R = Ce, Pr, Nd, Sm, Gd) single crystals were grown using metallic flux technique [18] with starting composition R:Au:Bi = 1:1:20. The mixture was placed in an alumina crucible and sealed in a quartz tube under vacuum. Then, they were taken to a conventional furnace and heated up to 1050°C with a rate of 50°C/h. After 4h at 1050°C, the batch was subjected to a slow cooling of 5°C/h down to 350°C. The excess Bi flux was then spun in a centrifuge and platelets-like crystals were mechanically removed from the crucible. The single crystals with dimensions 3.0 mm x 3.0 mm x 0.1 mm were grinded and their crystal structure and phase purity were determined by x-ray powder diffraction using a PANalytical diffractometer (Cu-Kα radiation). These compounds crystallize in a tetragonal $ZrCuSi_2$ structure (P4/nmm space group) and their lattice parameters are given in Table I. Elemental analysis of our single crystals using Energy Dispersive Spectroscopy (EDS) and Wavelength Dispersive spectroscopy (WDS) revealed the stoichiometry to be 1:0.92:1.6 for $CeAu_{0.92}Bi_2$[17] and very close to 1:1:2 for the others R = Pr, Nd, Gd, Sm with an error of 5%. Magnetic susceptibility measurements as function of the temperature were carried out using a commercial SQUID magnetometer. Specific-heat measurements were performed on a commercial small-mass calorimeter and electrical resistivity was measured using a low-frequency ac resistance bridge and four-contact configuration.

Table 1: Experimental parameters for $RAuBi_2$ (R = Ce, Pr, Nd, Sm, Gd)

	a	c	T_N	μ_{eff}	θ_p
	Å	Å	K	μ_B	K
$CeAuBi_2$[17]	4.647(5)	9.901(5)	12	2.55(2)	≈ - 21
$PrAuBi_2$	4.619(4)	9.861(8)	7.9	3.3(1)	≈ -19
$NdAuBi_2$	4.611(4)	9.816(8)	2.3	3.8(1)	≈ -43
$SmAuBi_2$	4.598(4)	9.765(8)	6.6		
$GdAuBi_2$	4.492(4)	10.045(8)	14.2	8.7(1)	≈ -58

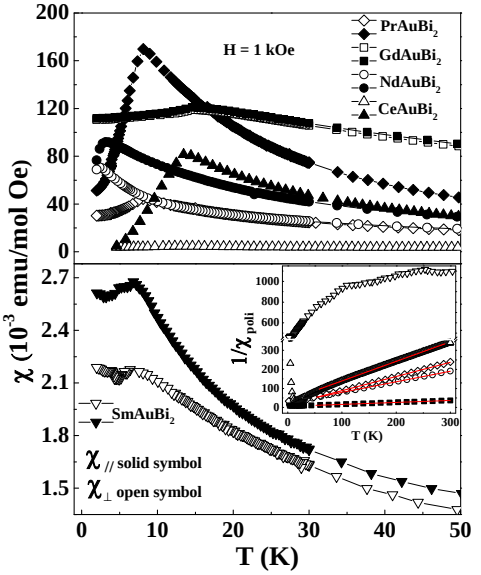

Figure 1: Temperature dependence of the magnetic susceptibility measured at $H = 1$ kOe applied along the c-axis,$\chi_{\parallel}$(solid symbols), and in the ab-plane,$\chi_{\perp}$(open symbols), for $RAuBi_2$ (R = Ce, Pr, Nd, Gd, Sm) materials. The inset shows the Curie-Weiss linear fits to χ^{-1} polycrystalline average.

3 Results and Discussion

The magnetic susceptibility as a function of temperature within the temperature range of 2 K $\lesssim$ T $\lesssim$ 50 K for a magnetic field $H = 1$ kOe applied along the c-axis, $\chi_{\parallel}$, and in the ab-plane, $\chi_{\perp}$, taken on single crystals of the studied compounds is shown in Figure 1. We found an antiferromagnetic (AFM) ordering at $T_N \sim 12.0$ K, 7.9 K, 2.3 K, 6.6 K and 14.2 K for $RAuBi_2$ (R = Ce, Pr, Nd, Sm, Gd), respectively. Furthermore, the data in fig.1 suggest a magnetic easy axis along the c-direction for all compounds, except for R = Gd. The inverse of the polycrystalline average of the magnetic susceptibility (χ^{-1}) is presented in inset of Figure 1. From Curie-Weiss linear fits to this averaged data in the range of 50-300K, we have extracted the Curie-Weiss temperature (θ_{CW}) and the effective magnetic moment (μ_{eff}) for all compounds, except for R = Sm. The obtained μ_{eff} were found to be close to their theoretical values (see Table I). In the case of the Sm-based compound, the high temperature magnetic anisotropy is probably associated with the CEF in both ground J=5/2 and excited J=7/2 multiplet states of the Sm^{3+} ion.[19] The non-Curie-Weiss magnetic susceptibility of $SmAuBi_2$ is commonly seen in Sm-based materials [19, 20, 21]. This behavior may be due to a Van Vleck contribution (which complicates the extraction of μ_{eff} and θ_{CW} from these data) [19].

The Figure 2 presents the magnetic specific heat divided by temperature $C_{mag}(T)/T$ in the temperature range 2 K $\lesssim$ T $\lesssim$ 25 K for the compounds of 1. To obtain the magnetic contribution to the specific heat, the phonon contribution was subtracted from the original data using the specific heat data of non-magnetic $LaAuBi_2$ compound. The peak-like anomaly in $C_{mag}(T)/T$ defines the AFM order that can be seen at $T_N \sim 12.0$ K, 7.9 K, 2.3 K, 6.6 K and 14.2 K, for $CeAuBi_2$, $PrAuBi_2$, $NdAuBi_2$, $SmAuBi_2$ and $GdAuBi_2$, respectively, [Fig. 2].

The Néel temperatures obtained from the specific heat data are in very good agreement with the magnetic susceptibility data (see Figure 1).

Figure 3 displays in-plane electrical resistivity ρ measured for all compounds. The high temperature data show a metallic behavior for these compounds. At low temperatures, subtle features can be seen at the respective ordering temperatures for some of the compounds.

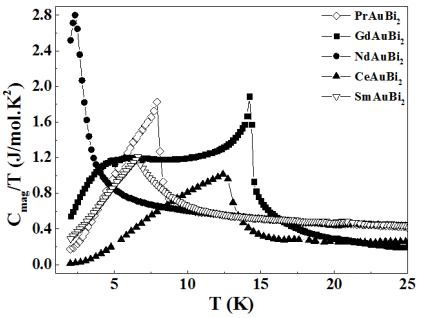

Figure 2: The magnetic contribution to the specific heat data divided by temperature in the temperature range 2 K $\lesssim$ T $\lesssim$ 25 K for $RAuBi_2$ (R = Ce, Pr, Nd, Gd, Sm).

In order to discuss the evolution of the magnetic properties in the $RAuBi_2$ family (R = Ce, Pr, Nd, Gd, Sm), we plot in Figure 4 the T_N values obtained for the studied compounds along the R-earth series, normalized by the its value for $GdAuBi_2$ (T_N = 14.2K).

The solid lines in Figure 4 are the de Gennes factor $[(g_J - 1)^2 J(J+1)]$ for ground state multiplet J through the rare-earths, where g_J is the Landé g-factor defined as $g_J = 3/2 + \{[S(S+1) - L(L+1]/J(J+1)\}$ and J is the total angular momentum. If one normalizes the de Gennes line to the $GdAuBi_2$ T_N value, for R = Sm and Nd the T_N values sit reasonably on the line but the T_N values for the R = Ce and Pr do not follow the de Gennes scaling. The actual values T_N for R = Ce and Pr are drastically above de Gennes lines. In fact, this effect is clearly present in other Ce-based and Pr-based intermetallic compounds [1, 20, 22, 23, 24, 21]. However, in the $RAuBi_2$ family the De Gennes breakdown of the T_N values for the Ce-based compounds is considerably larger in comparison with these examples found in literature [1, 20, 22, 23, 24, 21].

De Gennes scaling [25] neglects important contribution to the magnetic properties of these compounds such as CEF, Kondo effects and/or spatial dependence and anisotropic effects in the effective exchange parameter. Some of these effects are certainly expected to be present in the $RAuBi_2$ series. Therefore, it is not a surprise that the T_N values for complex rare-earth series in low-symmetry structure. Particularly, it has been shown for R_mMIn_{3m+2} (M = Rh, Ir and Co; m=1,2) complex classes of intermetallic compounds that the tetragonal CEF parameters strongly affects the RKKY interaction and determines both the direction of the ordered moments and the behavior of the ordering temperature for these series. [9, 26].

As such, further investigation and direct determination of the tetragonal CEF parameters along these series is in progress and it will be valuable to confirm the CEF effects in the antiferromagnetic interaction within the $RAuBi_2$ series. Furthermore, we will compare the results presented here with the properties of the isoestructural series $RCuBi_2$ [4, 17, 1] and discuss the main effects associated with the chemical substitution at the transition metal (Cu or Au) site. More broadly, detailed CEF analysis have been been of great importance to shed light on the evolution of the magnetic properties of the $CeNi_xBi_{2-y}$ [27] and $UAuBi_2$ [28] isoestructural compounds.

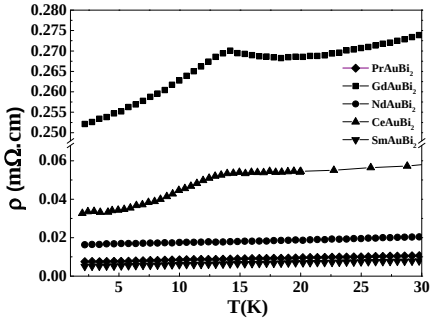

Figure 3: Temperature dependence of the electrical resistivity for $RAuBi_2$ (R = Ce, Pr, Nd, Gd, Sm) single-crystals.

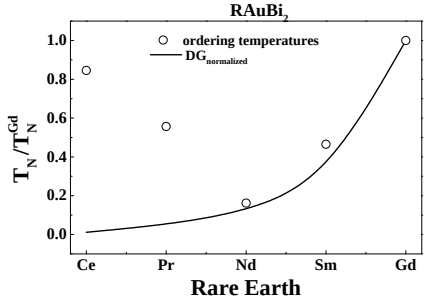

Figure 4: T_N values plotted of the compounds of the series $RAuBi_2$ (R = Ce, Pr, Nd, Gd, Sm) normalized to the T_N value for $GdAuBi_2$. The solid line is the de Gennes factor $[(g_J-1)^2 J(J+1)]$ for ground-state multiplet J of the rare earths normalized by the $GdAuBi_2$ T_N values.

4 Conclusions

In conclusion we report the evolution of the magnetic properties along the series of intermetallic compounds $RAuBi_2$ (R = Ce, Pr, Nd, Gd, Sm). Enlightened by our macroscopic measurements we found a dramatic breakdown of the De Gennes scaling in this series. We interpreted our data suggesting a CEF induced entrancement of T_N for the R = Ce and Pr compounds. This work was supported by FAPESP-SP (in particular Grants No 2013/17427-7, 2012/04870-7, 2006/60440-0, 2009/09247-3, 2010/11949-3, 2011/01564-0, 2011/23650-5, 2012/05903-6), CNPq, CAPES and FINEP-Brazil.

5 References

References

[1] C. B. R. Jesus, M. M. Piva, P. F. S. Rosa, C. Adriano, and P. G. Pagliuso, *Journal of Applied Physics* **115**, 17E115 (2014).

[2] J. G. S. Duque, C. Adriano, R. Lora-Serrano, C. Rettori, R. R. Urbano, J. L. Sarrao, S. B. Oseroff, and P. G. Pagliuso, *Journal of Applied Physics* **103**, 07B733 (2008).

[3] N. S. Sangeetha, A. Thamizhavel, C. V. Tomy, Saurabh Basu, S. Ramakrishnan, and D. Pal, *Physical Review B* **84**, 064430 (2011).

[4] C. Adriano, P. F. S. Rosa, C. B. R. Jesus, J. R. L. Mardegan, T. M. Garitezi, T. Grant, Z. Fisk, D. J. Garcia, A. P. Reyes, P. L. Kuhns, R. R. Urbano, C. Giles, and P. G. Pagliuso, *Physical Review B* **90**, 235120 (2014).

[5] M. H. Jung and A. H. Lacerda, *Physical Review B* **65**, 132405 (2002).

[6] A. Szytula, *Materials Science-Poland* **24**, 737 (2006).

[7] Hilbert v. Löhneysen, A. Rosch, M. Vojta, and Wölfle, *Rev. Mod. Phys.* **79**, 1015 (2007).

[8] J. D. Thompson, and Z. Fisk, *J. Phys. Soc. Japan* **81** 011002 (2012).

[9] P. G. Pagliuso, D. J. Garcia, E. Miranda, E. Granado, R. Lora-Serrano, C. Giles, J. G. S. Duque, R. R. Urbano, C. Rettori, J. D. Thompson, M. F. Hundley, and J. L. Sarrao, *J. Appl. Phys.* **99**, 08P703 (2006).

[10] A. D. Christianson, E. D. Bauer, J. M. Lawerence, P. S. Riseborough, N. O. Moreno, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, E. A. Goremychkin, F. R. Trouw, M. P. Hehlen, and R. J. McQueeney, *Phys. Rev. B* **70**, 134505 (2004).

[11] R. T. Macaluso, J. L. Sarrao, N. O. Moreno, P. G. Pagliuso, J. D. Thompson, F. R. Fronczek, M. F. Hundley, A. Malinowski, and J. Y. Chan, *Chemistry of Materials* **15**, 1394-1398 (2003).

[12] G. R. Stewart, *Rev. of Mod. Phys.* **73**, 797-855 (2001).

[13] G. R. Stewart, *Rev. of Mod. Phys.* **78**, 743-753 (2006).

[14] Genfu Chen, Shigeo Ohara, Masato Hedo, Yoshiya Uwatoko, Kazuya Saito, Michio Sorai, and Isao Sakamoto, *J. Phys. Soc. of Japan* **71**, 2836-2838 (2002).

[15] D. Kaczorowski, D. Gnida, A. P. Pikul, V.H. Tran, *Solid State Communications* **150**, 411-414 (2010).

[16] O. Stockert, J. Arndt, E. Faulhaber, C. Geibel, H. S. Jeevan, S. Kirchner, M. Loewenhaupt, K. Schmalzl, W. Schmidt, Q. Si, and F. Steglich *Nature Physics* **7**, 119 (2011).

[17] C. Adriano, P. F. S. Rosa, C. B. R. Jesus, T. Grant, Z. Fisk, D. J. Garcia, and P. G. Pagliuso, *Journal of Applied Physics* **117**, 17C103 (2015).

[18] Z. Fisk, J. P. Remeika, Handbook on the Physics and Chemistry of Rare Earth, **vol. 12**, Edited by: K. A. Gschneidner Jr. and L. Eyring, Amsterdam: Elsevier Science Publishers, 1989, p. 53.

[19] K. H. J. Buschow, H. W. de Wijn and A. M. Van Diepen, *J. Chem. Phys.* **50**, 137 (1969).

[20] C. Petrovic, S. L. Bud'ko, J. D. Strand, P. C. Canfield, *J. of Magnetism and Magn. Mat.* **261**, 210-221 (2003).

[21] Xiao Lin, Warren E. Straszheim, Sergey L. Bud'ko, P. C. Canfield, *J. of Alloys and Comp.* **554**, 304-311 (2013).

[22] K. D. Myers, S. L. Bud'ko, I. R. Fisher, Z. Islam, H. Kleinke, A.H. Lacerda, P.C. Canfield *J. of Magn. and Magn. Mat.* **205**, 27-52 (1999).

[23] P. G. Pagliuso, J. D. Thompson, M. F. Hundley, J. L. Sarrao, and Z. Fisk *Phys. Rev. B*, **63**, 054426 (2001).

[24] N. Van Hieu, T. Takeuchi, H. Shishido, C. Tonohiro, T. Yamada, H. Nakashima, K. Sugiyama, R. Settai, T. D. Matsuda, Y. Haga, M. Hagiwara, K. Kindo, S. Araki, Y. Nozue, and Y. Onuki, *J. Phys. Soc. of Japan*, **76**, 064702 (2007).

[25] P. G. De Gennes, *Compt. Rend.*, **247** (1958) and J. Physique Rad. **23**, 510 (1962); B. Coqblin, The Electronic Structure of Rare-Earth Metal and Alloys: the Magnetic Heavy Rare-Earths, Academic Press, New York, 1977).

[26] R. Lora-Serrano, D. J. Garcia, E. Miranda, C. Adriano, C. Giles, J. G. S. Duque and P. G. Pagliuso, *Phys. Rev. B* **79**, 024422 (2009).

[27] P. F. S. Rosa, C. B. R. Jesus, C. Adriano, Z. Fisk, and P. G. Pagliuso, *J. Phys.: Conf. Ser.* **592**, 012063 (2015).

[28] P. F. S. Rosa, Y. Luo, E. D. Bauer, J. D. Thompson, P. G. Pagliuso, and Z. Fisk, textitPhys. Rev. B **92**, 104425 (2015).