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Observation of a pressure-induced collapse of the Fe magnetic moment in the strong itinerant ferromagnet $Fe_{72}Pt_{28}$

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We observe a pressure-induced magnetic collapse from a ferromagnetic [high-moment (HM)] state to a low-moment (LM) magnetic state in the strong itinerant ferromagnet $Fe_{72}Pt_{28}$ (ordered and disordered phases). Such an existence of energetically closely lying HM and LM states demonstrates a unique physical picture of ferromagnetism in 3*d* magnetic systems that exhibit anomalously large magnetovolume effects. We further show that atomic ordering in $Fe_{72}Pt_{28}$ stabilizes the ferromagnetic ground state against pressure and modifies the spin correlations of the pressure-induced LM magnetic ground state.

The study of the pressure effect on the magnetic properties of Fe-based itinerant-electron ferromagnets such as the saturation magnetization M_s and the Curie temperature T_C is a fundamental issue in understanding the nature of ferromagnetism of 3*d* metals and alloys. This is related to the fact that an external pressure p varies the 3*d* correlations between magnetic neighboring atoms which results in a modification of the magnetic state of the system.[1] A strong mutual relationship between magnetism and volume has been found in a variety of Fe-based alloys and particularly in fcc ferromagnetic (FM) Fe-based Invar alloys.[2] Among these systems disordered fcc Fe-Ni Invar alloys have been considered as a model system to study itinerant electron ferromagnetism and thus have been the subject of numerous experimental and theoretical efforts.[3]

Many theoretical models have been proposed in order to explain the anomalously large magnetovolume effect (large decrease of M_s and T_C with pressure) and other magnetic anomalies in these alloys. Some of them are based on magnetic or compositional inhomogeneities in the framework of localized models.[4,5] Another approach is the itinerant electron model for very weak itinerant ferromagnets.[6] Both of these models give a good explanation of the magnetovolume effect in Fe-Ni Invar alloys. The validity of such models (see above), however, has been put in question with the discovery of Invar anomalies in fcc FM ordered Fe_3Pt and other Fe-Pt Invar alloys.[7] Fe_3Pt is found to be a *homogeneous strong* ferromagnet.[7] This result clearly shows that magnetic inhomogeneities and weak itinerant ferromagnetism are not necessary conditions for the magnetovolume effect in Invar alloys.

Recently, there has been renewed theoretical and experimental interest in the magnetic behavior of transition metals and alloys in connection with the Invar problem. On the theoretical side promising progress has been achieved by the investigation of the volume dependence of the FM Fe moment in pure fcc Fe using self-consistent spin-polarized band-structure calculations.[8,9] Here it is shown that the Fe magnetic moment in the FM [high-moment (HM) or high-spin[10]] state decreases sharply with decreasing volume, displaying a change of the magnetic order from HM to either a low-moment (LM) or low-spin[10] magnetic state or to a nonmagnetic (NM) state.[8,9] Regarding the experimental side, recent spin- and angle-resolved photoemission data on ordered Fe_3Pt at high temperatures ($T/T_C = 0.58$ to 1.3) have been reported as evidence for a high-spin to low-spin transition at *high* temperatures.[11] However, a very recent reexamination of these data on the basis of the single-site fluctuation theory shows that these data can be well fitted without assuming such a transition at high temperatures.[12]

In this paper we present a new approach towards a unique physical picture of the ferromagnetism in fcc Fe-based alloys and the related magnetovolume effect. We present studies of the stability of the FM state in ordered and disordered Fe_3Pt under high pressure and at low temperatures. Such experiments have not been reported so far. We observe for the *first* time in the strong itinerant ferromagnet $Fe_{72}Pt_{28}$ in both ordered and disordered phases a pressure-induced magnetic phase transition from the FM HM to a LM magnetic state at a critical pressure. The result is consistent with recent theoretical calculations of the volume dependence of the Fe magnetic moment in FM pure fcc Fe.[9] The existence of two closely lying magnetic states even in strong itinerant ferromagnets clearly demonstrates a unique physical picture of the ferromagnetism in 3*d* magnetic systems that exhibit magnetovolume anomalies.

We have investigated the magnetic properties of ordered and disordered $Fe_{72}Pt_{28}$ up to pressures of 8 GPa and at various temperatures (300–4.2 K) using the ^{57}Fe high-pressure Mössbauer-effect (ME) spectroscopy (^{57}Co: Rh source). The high-pressure setup is described elsewhere.[13] The ^{57}Fe ME technique is a powerful tool for the determination of pressure-induced changes of the Fe local magnetic moment $\mu_{Fe}(p)$ and of the Curie temperature $T_C(p)$. $\mu_{Fe}(p)$ is obtained by measuring the pressure dependence of the effective magnetic hyperfine (hf) field B_{eff} at the ^{57}Fe nucleus, since in a given Fe-alloy B_{eff} generally is proportional to μ_{Fe}. Such proportionality has been found to be particularly valid in the investigated Fe-Pt alloys.[7] $T_C(p)$ is obtained by measuring the temperature dependence of B_{eff} at different values of pressure.

The $Fe_{72}Pt_{28}$ samples were prepared by arc melting of 99.9985% pure Fe and 99.99% pure Pt. A proper amount of pure ^{57}Fe isotope (20%) was mixed with natural Fe in order to improve the ^{57}Fe Mössbauer resonance effect. Samples were homogenized at 1100 °C for about two weeks in an evacuated silica tube, then rolled down to thin foils of about 15 μm suitable for high-pressure measurements. The ordered ($T_C = 510$ K) and disordered ($T_C = 380$ K) $Fe_{72}Pt_{28}$ samples were obtained from the same ingot by a special thermal treatment described in Ref. 14. The purity and the stability of the fcc structure of the two samples down to 4.2 K were proved by x-ray diffraction experiments. We obtained values of the lattice constant a at 4.2 K of $a = 4.746(5)$ Å for the ordered sample and $a = 4.747(5)$ Å for the disordered sample. The long-range order parameter S (degree of order) was derived from the ratio of the integrated intensities of the fundamental and superstructure diffraction lines and according to the method given in Ref. 15. S was found to be 0 and 0.9 for the disordered and ordered samples, respectively.

Figure 1 shows some typical ME spectra of (a) disordered and (b) ordered $Fe_{72}Pt_{28}$ collected at 4.2 K and at different pressures. All spectra were fitted using a modified histogram method as described in Ref. 16. The fitting procedure allows one to obtain the value of the average hf field $\bar{B}_{\mathrm{eff}}$ and its distribution $P(B_{\mathrm{eff}})$. As expected, the relative width of $P(B_{\mathrm{eff}})$ in the ordered sample is found to be much smaller than that in the disordered sample. The dramatic decrease of the magnetic hf field in both phases at high pressures is clearly visible in Figs. 1(a) and 1(b). We observe in both phases a reversible behavior upon releasing pressure. Figure 2 shows the variation of the relative decrease of $\bar{B}_{\mathrm{eff}}$, $\bar{B}_{\mathrm{eff}}(p)/\bar{B}_{\mathrm{eff}}(0)$, and of T_C, $T_C(p)/T_C(0)$, for both $Fe_{72}Pt_{28}$ samples. We observe in the two phases a sharp decrease of both physical quantities [$\bar{B}_{\mathrm{eff}}(p)$ and particularly $T_C(p)$] at a critical pressure p_c [see Figs. 2(a) and 2(b)]. This clearly indicates a pressure-induced magnetic phase transition at p_c 4.2 GPa for the disordered phase and at $p_c \simeq 6$ GPa for the ordered phase. The discontinuous decrease of T_C in both samples at p_c is more pronounced that that of $\bar{B}_{\mathrm{eff}}$. This is due to the fact that $\bar{B}_{\mathrm{eff}}$ is an average value of the distribution of the Fe local magnetic moment in the two samples (see below). We attribute this magnetic phase transition at p_c to a transition from a FM HM state to a LM magnetic state as recently predicted from theoretical calculations on fcc FM Fe.[9] The evolution of the HM to LM transition with increasing pressure is best seen in the change of the shape of the hf field distribution P (B_{eff}) with pressure in the disordered phase [see Fig. 3(a)]. Here, the pressure-induced population of the LM state increases at the expense of that of the HM state, while no shift occurs from the HM state towards the LM state. The same feature is observed in the ordered phase. Thus, such an evolution of the pressure dependence of the magnetic ground state in $Fe_{72}Pt_{28}$ reveals the existence of two *discrete* closely lying magnetic states as expected from spin-polarized band-structure calculations on fcc Fe.[9] This new experimental finding should stimulate theoreticians to perform such calculations on $Fe_{72}Pt_{28}$.

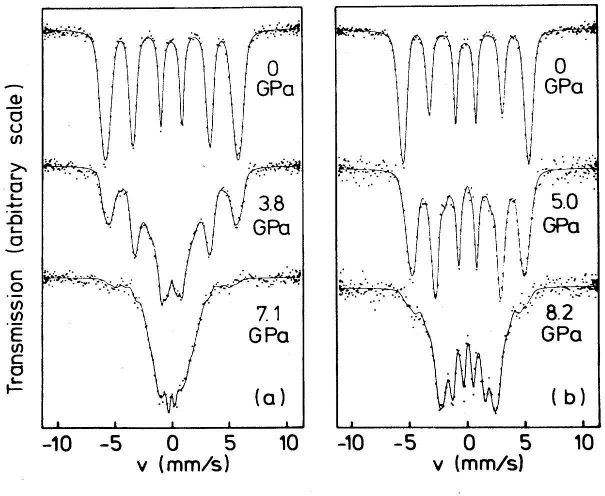

FIG. 1. ^{57}Fe Mössbauer spectra of $Fe_{72}Pt_{28}$ at 4.2 K and various pressures: (a) disordered phase, (b) ordered phase. Solid lines give fit to data as discussed in text.

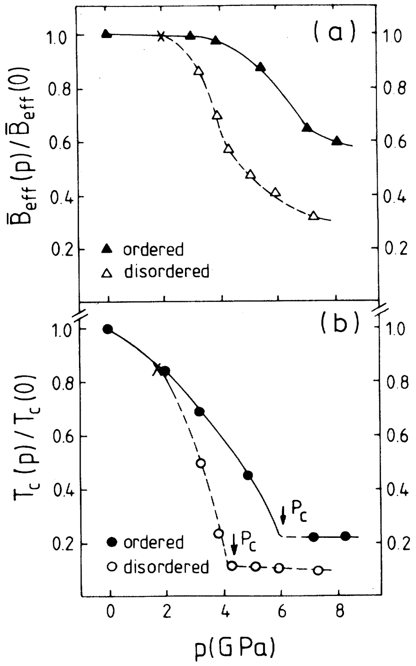

FIG. 2. (a) Pressure dependence of the average effective hf field $\bar{B}_{\mathrm{eff}}$ and (b) the Curie temperature T_C for disordered and order phase of $Fe_{72}Pt_{28}$. P_c marks the critical pressure at which the HM to LM phase transition occurs. Data points at 1.8 GPa (×) are taken from Ref. 17. Lines through data points are only a guide to the eye.

In order to gain a deeper insight into the nature of itinerant ferromagnetism in $Fe_{72}Pt_{28}$ we compare in Fig. 4 the pressure data on $Fe_{72}Pt_{28}$ with those for the weak itinerant ferromagnet $Fe_{68.5}Ni_{31.5}$ (from Ref. 18) and with theoretical calculations on pure fcc Fe (from Ref. 9). Here the relative decrease of the average Fe magnetic mo-

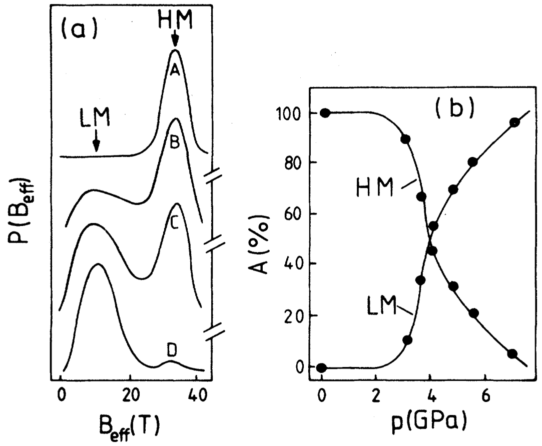

FIG. 3. (a) Magnetic hf field distribution $P(B_{\mathrm{eff}})$ of disordered $Fe_{72}Pt_{28}$ at 4.2 K and at various pressures as obtained from least-squares fits to the corresponding ME spectra: A (0 GPa); B (3.8 GPa); C (5.0 GPa), and D (7.1 GPa). (b) Fraction A of HM and LM state in disordered $Fe_{72}Pt_{28}$ at various pressures.

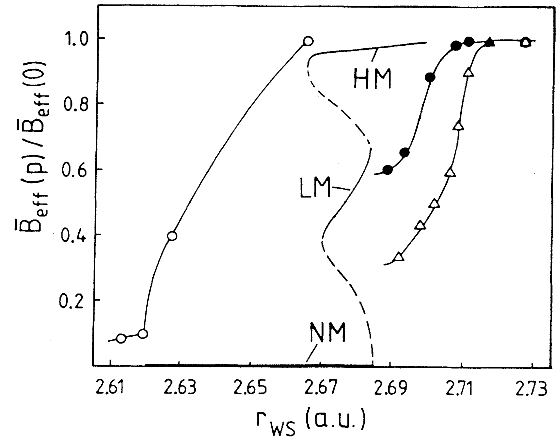

FIG. 4. Measured values of $\bar{B}_{\mathrm{eff}}(0)$ as a function of the Wigner-Seitz radius r_{WS} of the Fe WS sphere for $Fe_{72}Pt_{28}$ ordered (●) and disordered (△) and $Fe_{68.5}Ni_{31.5}$ (○) (from Ref. 18); data point (▲) is taken from Ref. 17. Solid lines through data points are only a guide to the eye. Additionally shown is the solid-dashed theoretical curve for $\mu_{Fe}(p)/\mu_{Fe}(0)$ in fcc Fe (from Ref. 9). The dashed parts of the theoretical curve indicate metamagnetic regions.

ment $\bar{B}_{\mathrm{eff}}(p)/\bar{B}_{\mathrm{eff}}(0)$ is plotted versus the estimated valued of the Wigner-Seitz (WS) radius (r_{WS}) of the Fe WS sphere in fcc Fe, $Fe_{72}Pt_{28}$ (ordered and disordered) and disordered $Fe_{68.5}Ni_{31.5}$. Figure 4 shows a *unique* behavior of the volume dependence of μ_{Fe} of $Fe_{72}Pt_{28}$, $Fe_{68.5}Ni_{31.5}$, and pure fcc Fe.[9] In all systems one finds a pressure-induced magnetic phase transition from a HM to a LM magnetic state at different critical values of r_{WS}. Such differences in r_{WS} should be related to differences in the band structure and the exchange interactions involved in each of the systems. On the other hand, the *initial* rate of decrease of μ_{Fe} and T_C for $0 \leq p \lesssim 2$ GPa are entirely different in disordered $Fe_{72}Pt_{28}$ and disordered $Fe_{68.5}Ni_{31.5}$: in weak itinerant $Fe_{68.5}Ni_{31.5}$, the initial rate of decrease of μ_{Fe} is nearly equal to that of T_C (Ref. 18) as expected from the theoretical model for weak itinerant ferromagnets.[6] Contrary to this we obtain in strong itinerant $Fe_{72}Pt_{28}$ [see Figs. 2(a) and 2(b)] no change of μ_{Fe} despite the rapid decrease of T_C with pressure ($dT_C/dp = -38$ K GPa^{-1}). Thus, we relate the different pressure-induced decrease of μ_{Fe} ($0 \leq p \lesssim 2$ GPa) in the two systems to a highly localized magnetic moment in $Fe_{72}Pt_{28}$ compared to $Fe_{68.5}Ni_{31.5}$. However, as the pressure increases $2 \lesssim p \lesssim 4$ GPa, the highly localized character of μ_{Fe} in $Fe_{72}Pt_{28}$ disappears and the system behaves as a *weak* itinerant ferromagnet. The rate of decrease of T_C and μ_{Fe} with pressure is nearly equal [see Figs. 2(a) and 2(b)], similar to those in weak itinerant $Fe_{68.5}Ni_{31.5}$ (see above).

Next we discuss another interesting aspect of our studies in $Fe_{72}Pt_{28}$, namely the effect of *atomic* ordering on the stability of the FM ground state under high pressure. As evident from Fig. 2 (see also Fig. 4), the magnetic ground state in the ordered phase is more stable against pressure than the disordered phase. The ordered phase exhibits a smaller rate of decrease of μ_{Fe} and T_C with pressure and a higher value of the critical pressure when compared with the disordered phase (see Fig. 2). A higher stability of the FM ground state in the ordered phase against pressure is consistent with its higher value of T_C with respect to that of the disordered phase. On the other hand, we have recently shown that the pressure-induced instability of the FM state in fcc, Fe-based Invar alloys is enhanced with increasing the average number (n) of Fe nearest neighbors (nn) or the average number of Fe pairs.[18,19] On this basis it can be easily shown that atomic ordering in $Fe_{72}Pt_{28}$ reduces the average number of Fe nn with respect to the disordered phase and thereby increases the stability of the FM state of the ordered phase against pressure: one obtains $n = 8$ for perfectly ordered $Fe_{72}Pt_{28}$ phase (Cu_3Au-type superlattice structure) and $n = 8.64$ for the corresponding disordered phase.

Finally, we want to stress on an important consequence of atomic ordering in $Fe_{72}Pt_{28}$. As evident from Figs. 2(a) and 2(b), we find (well above p_c) two different pressure-induced LM magnetic ground states in the two phases with respect to the values of $T_C(p)$ and $\bar{B}_{\mathrm{eff}}(p)$. We obtain for the ordered phase values of $T_C = 100$ K and $\bar{B}_{\mathrm{eff}} = 16$ T at $p = 7.1$ GPa and for the disordered phase values of $T_C = 40$ K and $\bar{B}_{\mathrm{eff}} = 10$ T at $p = 8.2$ GPa. Despite the fact that this finding cannot prove the *type* of magnetic order in the two phases, it clearly shows that the local spin correlation of the Fe moments in the two phases are different as a consequence of different atomic ordering.

In conclusion, our observation of a pressure-induced collapse of Fe moment in the strong itinerant ferromagnet $Fe_{72}Pt_{28}$ reveals a unique physical picture of ferromagnetism in fcc $3d$ magnetic systems that exhibit anomalously large magnetovolume effects. Also, we feel that our high pressure results may stimulate theoretical efforts concerning the pressure dependence of μ_{Fe} in Fe_3Pt as well the effect of atomic ordering on the stability of its FM ground state.

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[1] J. S. Schilling, Adv. Phys. **28**, 657 (1979).

[2] Y. Nakamura, IEEE Trans. Magn. **MAG-12**, 278 (1976).

[3] See, for example, *The Invar Problem,* edited by A. J. Freeman and M. Shimizu (North-Holland, Amsterdam, 1979).

[4] E. I. Kondorskii and V. L. Sedov, J. Appl. Phys. Suppl. **31**, 331 (1960).

[5] R. J. Weiss, Proc. Phys. Soc. **82**, 281 (1963).

[6] D. Wagner and E. P. Wohlfarth, J. Phys. F **11**, 2417 (1981), and references therein.

[7] Y. Nakamura, K. Sumiyama, and M. Shiga, J. Magn. Magn. Mater. **12**, 127 (1979).

[8] C. S. Wang, B. M. Klein, and H. Krakauer, Phys. Rev. Lett. **54**, 1852 (1985).

[9] V. L. Moruzzi, P. M. Marcus, K. Schwarz, and P. Mohn, Phys. Rev. B **34**, 1784 (1986).

[10] The terms high-spin and low-spin states have been commonly used in the literature (e.g., Refs. 9, 11, and 12). Since these terms originally used to describe electronic configurations of Fe in insulating systems with localized moment, we prefer to use the terms high-moment and low-moment states.

[11] E. Kisker, E. F. Wassermann, and C. Carbone, Phys. Rev. Lett. **58**, 1784 (1987).

[12] Y. Kakehashi, Phys. Rev. B **38**, 12051 (1988).

[13] M. M. Abd-Elmeguid, H. Micklitz, and I. Vincze, Phys. Rev. B **25**, 1 (1982).

[14] H. Maruyama, J. Phys. Soc. Jpn. **55**, 2834 (1986).

[15] J. M. Cowley, Phys. Rev. **77**, 669 (1950).

[16] R. A. Brand, J. Lauer, and D. M. Herlach, J. Phys. F **13**, 675 (1983).

[17] K. Hayashi and N. Mori, Solid State Commun. **38**, 1057 (1981).

[18] M. M. Abd-Elmeguid, B. Schleede, and H. Micklitz, J. Magn. Magn. Mater. **72**, 253 (1988).

[19] W. Ludorf, M. M. Abd-Elmeguid, and H. Micklitz, J. Magn. Magn. Mater. **78**, 171 (1989).