SUPPLEMENTAL MATERIAL

Experimental evidences for static charge density waves in iron oxy-pnictides

S.1: Synchrotron powder diffraction

The structural properties of La($Fe_{1-x}Mn_x$)AsO samples (x = 0.02 and 0.04) were investigated by means of high-resolution synchrotron X-ray powder diffraction analysis between 290 K and 10 K; X-ray scattering data were collected at the ID22 high-resolution powder diffraction beamline of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. Structural data at 290 K and 10 K obtained after Rietveld refinement are listed in Tables S1 and S2.

Table S1: Structural data at 290 K obtained for La($Fe_{1-x}Mn_x$)AsO samples after Rietveld refinement in the *P*4/*nmm* space group (synchrotron X-ray diffraction data).

		La($Fe_{0.98}Mn_{0.02}$)AsO			La($Fe_{0.96}Mn_{0.04}$)AsO		
Cell parameters	*a* (Å)	4.0372(1)			4.0391(1)		
	c (Å)	8.7505(1)			8.7640(1)		
Atomic site		*x*	*y*	*z*	*x*	*y*	*z*
La	2*c*	¼	¼	0.1411(1)	¼	¼	0.1409(1)
(Fe,Mn)	2*a*	¾	¼	½	¾	¼	½
As	2*c*	¼	¼	0.6518(1)	¼	¼	0.6521(1)
O	2*b*	¾	¼	0	¾	¼	0
R_F (%)		4.50			3.38		
R_{Bragg} (%)		8.75			2.97		

Table S2: Structural data at 10 K obtained for La($Fe_{1-x}Mn_x$)AsO samples after Rietveld refinement in the *Cmme* space group (synchrotron X-ray diffraction data).

		La(Fe$_{0.98}$Mn$_{0.02}$)AsO			La(Fe$_{0.96}$Mn$_{0.04}$)AsO		
Cell parameters	*a* (Å)	5.7066(1)			5.7066(1)		
	b (Å)	5.6911(1)			5.7066(1)		
	c (Å)	8.7206(1)			8.7310(1)		
Atomic site		*x*	*y*	*z*	*x*	*y*	*z*
La	4*g*	0	¼	0.1417(1)	0	¼	0.1415(1)
(Fe,Mn)	4*b*	¼	0	½	¼	0	½
As	4*g*	0	¼	0.6511(1)	0	¼	0.6514(1)
O	4*a*	¼	0	0	¼	0	0
R_F (%)			5.43			4.97	
R_{Bragg} (%)			4.73			4.90	

Generally speaking, the variation of the lattice parameters of incommensurately modulated structures may induce microstrain-like anisotropic line broadening in powder diffractions patterns [1]. In this context, Figure S1 shows the superposition of the Williamson-Hall plots obtained after Rietveld refinements of the data collected at 10 K and 290 K (x = 0.02); for a sake of clarity, data at 10 K are reported according to the tetragonal setting. It is evident that low temperature data are affected by a micro-strain like contribution to the line broadening. From these data it can be concluded that microstrain increases by almost 30% along [00*l*] and <*h*00>, but the gain exceeds 50% along <*hh*0>, which represents the direction of the Fermi surface nesting wave vector.

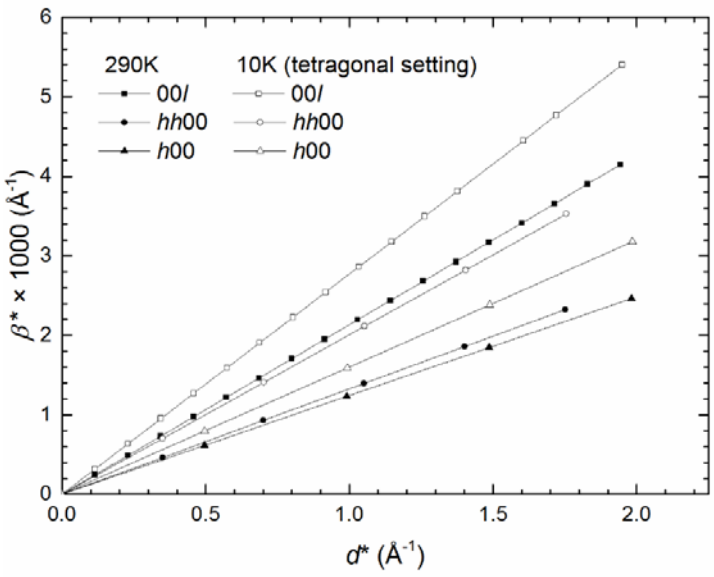

Figure S1: Superposition of the Williamson-Hall plots obtained after Rietveld refinements of the data collected at 10 K and 290 K for the La(Fe$_{0.98}$Mn$_{0.02}$)AsO sample.

This behaviour complies with the case where only the parameters describing the average lattice vary in the different crystallites, but not the modulation vector[1]. This is confirmed by the fact that the position of the satellite reflection remains constant with temperature.

S.2: Determination of the magnetic transition temperature

Neutron diffraction analysis is the best and direct probe of a magnetically ordered state. Preliminary data were collected for the $La(Fe_{0.98}Mn_{0.02})AsO$ sample using the high-intensity medium-resolution neutron powder D20 diffractometer of the Institute Laue-Langevin (λ = 2.41 Å). Remarkably, by these neutron diffraction data, also the structural transition temperature can be determined to T_s = 113.5±0.5 K, in optimal agreement with the result obtained by synchrotron X-ray powder diffraction data. Figure S2 shows the thermal evolution of the magnetic moment as obtained after Rietveld refinement using the high-intensity neutron powder diffraction data; a T_{SDW} = 99.5±0.5 K results by fitting these data using a Landau mean field model $m(T) = m_0(1 - T/T_m)^{\beta}$, where m_0 is the magnetic moment extrapolated at T = 0 K and β is the critical exponent, which is ½ in the case of pure 2nd order transitions. This result complies with the magnetization measurement; Figure S.3 shows the magnetization curve and the corresponding *erf* fit; the inflection point is located at 104.5±0.4 K, a few degrees above the long range AF order probed by neutrons.

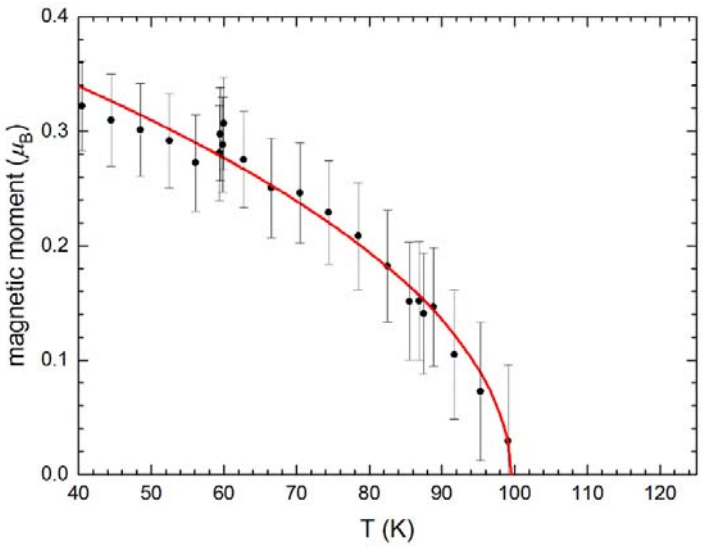

Figure S2: Thermal evolution of the magnetic moment in the $La(Fe_{0.98}Mn_{0.02})AsO$ sample as obtained after Rietveld refinement of high-intensity low-resolution neutron powder diffraction data; the data are fitted according to a Landau mean field theory valid for a 2nd order transition (red line).

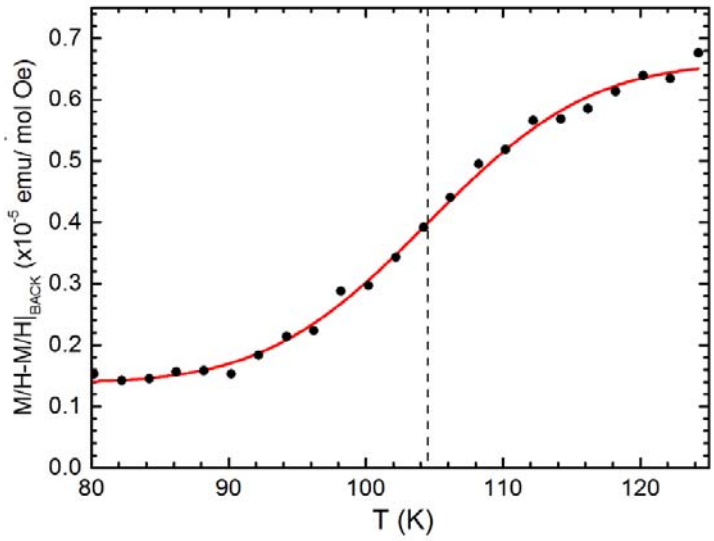

Figure S3: Temperature dependence of M/H measured under an applied field of 3 T in the magnetic transition region after subtracting a Curie–Weiss-like 'background' ($M/H|_{BACK}$) [2]; the red line represents the *erf*-fit whose inflection point is at $T \sim 104.5$ K (dashed line).

S.3: dc resistivity

Figure S4 shows the electrical resistivity (normalized to its value at T = 300 K) measured as a function of the temperature. The increase of the Mn content causes a suppression of structural/magnetic ordering temperature with a progressive smoothing of the typical step-like feature that characterizes the $x = 0$ composition around $T = 150$ K (blue curve in Figure S4). Moreover, the substitution of even a small amount of Mn substantially changes the nature of conduction. In fact in the $x = 0.01$ sample, the resistivity which decreases below the magnetic/structural transition, undergoes an abrupt upturn below 50 K. In the heavier substituted samples ($x = 0.02$ and 0.04) the low temperature upturn appears superimposed to a semiconducting-like behaviour starting at room temperature.

The inset of Figure S4 shows the evolution of the Hall coefficient curves, R_H vs T, as a function of the Mn content. The increase of the Mn content causes a suppression of the structural/magnetic ordering temperature and a progressive diminution of the absolute value, which can be ascribed to a compensation effect characteristic of a multiband system.

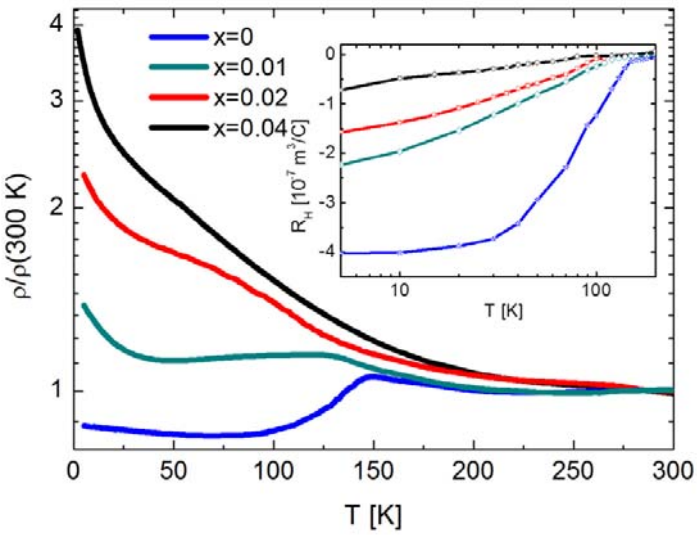

Figure S4: Normalized resistivity ρ/ρ(300 K) vs T measurement of $La(Fe_{1-x}Mn_x)AsO$ (x =0, 0.01, 0.02 and 0.04) series; inset: Hall coefficient R_H vs T measurement of $La(Fe_{1-x}Mn_x)AsO$ (x =0, 0.01, 0.02 and 0.04) series.

References

[1] A. Leineweber, V. Petricek, Microstrain-like diffraction-line broadening as exhibited by incommensurate phases in powder diffraction patterns, J. Appl. Cryst. **40**, 1027 (2007)

[2] McGuire, M. A., Hermann, R.P., Sefat, A. S., Sales, B.C., Jin, R., Mandrus, D., Grandjean, F., Long, G. J., New J. Phys. **11**, 025011 (2009)