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A Mössbauer study of $Ba(Fe_{1-x}Mn_x)_2As_2$ single crystals

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Abstract Single crystals of $Ba(Fe_{1-x}Mn_x)_2As_2$ (x = 0, 0.016, 0.064) have been prepared and studied by ^{57}Fe Mössbauer spectroscopy in the temperature range from 30 K to room temperature. The Mössbauer spectra show a change in the spectral shape with the change of Mn concentrations. It is found that both the hyperfine field and the magnetic phase transition temperature decrease with the increasing of Mn concentrations.

Keywords Magnetic phase transition · Mössbauer spectroscopy · $Ba(Fe_{1-x}Mn_x)_2As_2$

1 Introduction

Recently, systematic studies of underdoped $BaFe_2As_2$ have revealed interesting results regarding the relationship between structure, magnetism and superconductivity. With the decreasing of temperature, the $BaFe_2As_2$ parent compound undergoes simultaneous structural and magnetic phase transitions from a paramagnetic tetragonal phase to an orthorhombic antiferromagnetic phase at 140 K [1]. The structural/magnetic phase transition appears to split into two distinct phase transitions, both of which are suppressed with increasing electron doping (Co or Ni) at Fe site, and superconductivity arises for an appreciable range of doping [2, 3]. On the contrary, hole doping by Cr or Mn at the Fe site does not induce superconductivity, although the structural and magnetic phase transition are also suppressed [4, 5].

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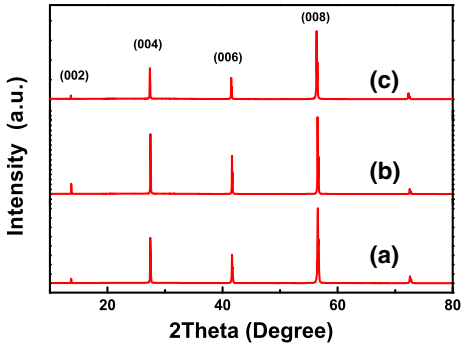

Fig. 1 XRD patterns of the prepared single crystals of $Ba(Fe_{1-x}Mn_x)_2As_2$: **a** x = 0, **b** 0.016, **c** 0.064

Given the strong coupling between structure, magnetism and superconductivity, the magnetic and structural properties of the hole doped $BaFe_2As_2$ compounds also require further investigations.

In addition, in iron-based superconductors magnetism, as well as superconductivity, is confined to Fe-As layers. Further investigations have shown that in Ba-122 iron pnictide, the Fe sublattice shows a spin density wave (SDW) magnetic order [1, 6]. Therefore, the ^{57}Fe Mössbauer spectroscopy has a natural advantage in studying the hyperfine fields at the Fe nuclei and the related magnetic properties of Fe atoms. There have been some reports about the observation of the SDW magnetic order using Mössbauer spectroscopy [6–9]. In this paper, we present Mössbauer study of powdered single crystals of $Ba(Fe_{1-x}Mn_x)_2As_2$ with x = 0, 0.016, 0.064.

2 Experimental details

Single crystals of $Ba(Fe_{1-x}Mn_x)_2As_2$ (x = 0, 0.016, 0.064) were grown out of a FeAs self-flux using conventional high-temperature solution growth techniques [2]. The prepared samples were well-formed plates with typical size of 5 × 5 × 0.2 mm^3 and the actual Mn concentrations were determined by the atomic absorption spectroscopy. The X-ray diffraction results, as shown in Fig. 1, suggest that the crystallographic c-axis is perpendicular to the plane of the plate. Transmission Mössbauer spectroscopy studies were performed using a conventional constant acceleration drive with a ^{57}Co:Pd source. The absorbers were prepared in powder form (10 mg of natural Fe/cm^2). The velocity calibration was performed with sodium nitroprusside powder at room temperature (RT) and all isomer shifts were quoted relative to the α-Fe foil at RT.

3 Results and discussion

Mössbauer spectra of $Ba(Fe_{1-x}Mn_x)_2As_2$ (x = 0, 0.016, 0.064) single crystals at temperatures between 30 K and RT are shown in Fig. 2. At RT, the spectra are single absorption lines with a little asymmetry, indicating a small amount of a foreign FeAs phase [10]. The content of the FeAs phase is too small to be reasonably fitted, so the spectra were fitted with single lines. As can be seen, at temperatures below the

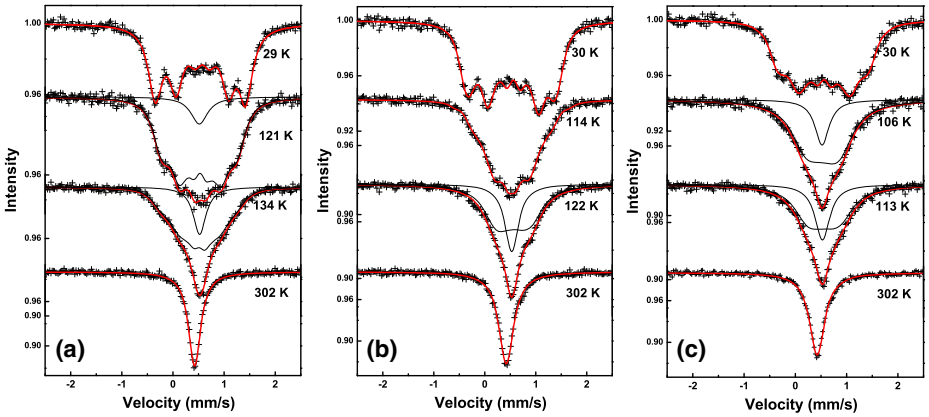

Fig. 2 ^{57}Fe Mössbauer spectra of the single crystals of $Ba(Fe_{1-x}Mn_x)_2As_2$ obtained at indicated temperatures: **a** $BaFe_2As_2$; **b** $Ba(Fe_{0.984}Mn_{0.016})_2As_2$; **c** $Ba(Fe_{0.936}Mn_{0.064})_2As_2$

Table 1 Selected hyperfine parameters as obtained for $Ba(Fe_{1-x}Mn_x)_2As_2$

x	Temperature (K)	IS (mm/s)	HF (T)	2ε (mm/s)
0	29	0.56	5.24	−0.05
	RT	0.42	–	–
0.016	30	0.54	4.79	−0.04
	RT	0.42	–	–
0.064	30	0.55	4.49	−0.03
	RT	0.43	–	–

IS is the isomer shift, HF is the average hyperfine field and 2ε is the quadrupole shift

SDW transition point, the spectra are not typical sextets and it is impossible to fit the data with only one sextet. Therefore, the spectra were fitted assuming a distribution of hyperfine fields. The hyperfine parameters at selected temperatures are listed in Table 1.

As shown in Table 1, the isomer shift (IS) values for all samples are around 0.42 mm/s at RT, which is typical for the $Th_2Cr_2Si_2$-type compounds. The almost identical IS values may be due to the small doping level of the samples or the fact that doping at iron site by similar atoms, such as Co, Ni, has little effect on IS [8]. The hyperfine fields of $Ba(Fe_{1-x}Mn_x)_2As_2$ exhibit strong dependence on the Mn concentrations. First, the hyperfine field drops from 5.24 T for $BaFe_2As_2$ to 4.79 T and 4.49 T for $x = 0.016$ and $x = 0.064$ around 30 K, respectively. Second, as shown in Fig. 2a, the spectrum of the parent compound changes from a singlet pattern to a complex pattern with magnetic splitting around 134 K, and the transition temperature is similar to the reported results [1, 10]. However, for the Mn-containing samples, the magnetic transitions are suppressed to lower temperatures with the increasing of Mn concentrations: 122 K for $x = 0.016$ and 113 K for $x = 0.064$. Third, as shown in Fig. 2, the linewidths of the spectra obtained around 30 K increase obviously for the doped compounds, which has also been observed in $Ba(Fe_{1-x}Ni_x)_2As_2$ system [9]. The broadening of the linewidth suggests a distribution of the local magnetic environment, which may be caused by the coexistence of commensurate and incommensurate SDWs [9, 11] and/or inhomogeneous Mn substitutions.

4 Conclusion

In conclusion, Mössbauer studies of single crystals of $Ba(Fe_{1-x}Mn_x)_2As_2$ have been performed. Our results show that Mn-substitution changes the shape of the spectra significantly and both the hyperfine field and the SDW transition temperature decrease with the increase of the Mn concentrations in the range of $0 < x < 0.064$.

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