# Phase separation in superconducting and antiferromagnetic $Rb_{0.8}Fe_{1.6}Se_2$ probed by Mössbauer spectroscopy

Vadim Ksenofontov,[1,*] Gerhard Wortmann,[2] Sergey A. Medvedev,[1,3] Vladimir Tsurkan,[4,5] Joachim Deisenhofer,[4] Alois Loidl,[4] and Claudia Felser[1]

[1]*Institut für Anorganische und Analytische Chemie, Johannes Gutenberg-Universität, D-55099 Mainz, Germany*
[2]*Department Physik, Universität Paderborn, D-33095 Paderborn, Germany*
[3]*Max-Planck Institute for Chemistry, D-55128 Mainz, Germany*
[4]*Experimental Physics V, University of Augsburg, D-86159 Augsburg, Germany*
[5]*Institute of Applied Physics, Academy of Sciences, MD-2028, Chişinău, Republic of Moldova*
(Received 15 August 2011; revised manuscript received 23 October 2011; published 15 November 2011)

$^{57}$Fe-Mössbauer studies of superconducting $Rb_{0.8}Fe_{1.6}Se_{2.0}$ with $T_C = 32.4$ K were performed on single-crystalline and polycrystalline samples in the temperature range 4.2–295 K. They reveal the presence of 88% magnetic and 12% nonmagnetic $Fe^{2+}$ species with the same polarization dependence of their hyperfine spectra. The magnetic species are attributed to the $16i$ sites of the $\sqrt{5} \times \sqrt{5} \times 1$ superstructure and the nonmagnetic Fe species to a nanosized phase observed in recent structural studies of superconducting $K_xFe_{2-y}Se_2$ systems rather than to the vacant $4d$ sites in the $\sqrt{5} \times \sqrt{5} \times 1$ superstructure. The $^{57}$Fe spectrum of a single-crystalline sample in an external field of 50 kOe applied parallel to the crystallographic $c$ axis confirms the antiferromagnetic order between the fourfold ferromagnetic Fe($16i$) supermoments and the absence of a magnetic moment at the Fe sites in the minority phase. A discussion of all spectral information and comparison with superconducting FeSe provides convincing evidence that the nanoscale phase separation is monitored by Mössbauer spectroscopy in $Rb_{0.8}Fe_{1.6}Se_{2.0}$.

Fe-based superconductors are at the center of recent interest since superconducting (SC) transition temperatures $T_C$ up to 55 K were observed.[1] These superconductors all have the same structural motif, namely, Fe-$X$ layers, where $X$ is either As or Se, both also modified by substitution. Superconducting FeSe and related Te-substituted systems possess the simplest structure, consisting of Fe-Se layers only. FeSe exhibits a relatively low $T_C$ value of 8.5 K, however, a significant increase of $T_C$ up to 36.7 K is observed under a pressure of ∼9 GPa.[2–4] At present, the microscopic mechanisms of superconductivity in these Fe-based superconductors are widely discussed, e.g., as based on spin fluctuations or complex interactions between occupied and nonoccupied structures at the Fermi edge of the valence band, built up dominantly from Fe $3d$-band electrons.[5] The observation of superconductivity in magnetic members of the $SrFe_2As_2$ family called into question the old paradigm that superconductivity cannot coexist with magnetism,[5] similar to previous observations in heavy-fermion superconductors.[6] This actual discussion was stimulated by the reports of coexistence of superconductivity and magnetism in another family of Fe-based superconductors, namely, the $A_xFe_{2-y}Se_2$ ($A$ = K, Rb, Cs) systems exhibiting $T_C$ values up to 32 K and antiferromagnetic ordering temperatures above 500 K, both properties representing bulk samples.[7,8] These compounds are closely related to superconducting, but nonmagnetic FeSe, but now with the Fe-Se layers separated by alkali ions. In this respect they are similar to the above-mentioned $SrFe_2As_2$ systems with $ThCr_2Si_2$ structure, however, with an important difference with a $\sqrt{5} \times \sqrt{5} \times 1$ superstructure modulating the Fe deficiency in the Fe-Se layers by the formation of Fe ions in $16i$ positions and of empty Fe sites in $4d$ positions.[9,10] Very recent reports about a nanosized filamentary phase formed concomitantly with the main $\sqrt{5} \times \sqrt{5} \times 1$ super-structure have provided further aspects about the coexistence of superconductivity and magnetism in $K_xFe_{2-y}Se_2$ systems. Yuan *et al.*[11] reported on a nanoscale phase separation of the magnetic $\sqrt{5} \times \sqrt{5} \times 1$ superstructure together with a minority K-deficient superconducting phase in $K_{0.75}Fe_{1.75}Se_2$. Ricci *et al.*[12] reported also on a nanoscale phase separation in $K_{0.8}Fe_{1.6}Se_2$ and suggested that the coexistence of insulating vacancy-ordered magnetic domains with a $\sqrt{5} \times \sqrt{5} \times 1$ structure and metallic nonmagnetic domains is responsible for superconductivity.

In this Rapid Communication we provide strong evidence for this phase separation in a well-characterized superconducting single-crystalline $Rb_{0.8}Fe_{1.6}Se_{2.0}$ sample investigated by $^{57}$Fe-Mössbauer spectroscopy, which is especially suited because of site-specific information on the local magnetic, electronic, and structural properties of the Fe species, which are the active players for the magnetic and superconducting properties.

Single crystals of the Rb-Fe-Se system were grown by the Bridgman method using as starting materials the elemental Rb (99.75%) and high-purity FeSe previously synthesized by solid-state reactions.[13] Plateletlike single crystals with diameters of ∼10 mm were separated from the solidified melt. X-ray diffraction (XRD) studies at 295 K on a powdered sample, grinded from small crystals under inert conditions, revealed lattice parameters $a = 3.9228(7)$ Å and $c = 14.5909(38)$ Å assuming $I4/mmm$ symmetry. The composition of the sample was determined by wavelength-dispersive electron-probe microanalysis by averaging over 30 spots on the surface of a freshly cleaved single-crystalline sample as $Rb_{0.80(3)}Fe_{1.60(1)}Se_{2.00(4)}$, hereafter referred to as $Rb_{0.8}Fe_{1.6}Se_2$. For all details about sample preparation and characterization, see Ref. 13.

Magnetic susceptibility measurements delivering the superconducting and magnetic properties were performed in the temperature range 2–600 K and in magnetic fields up to 50 kOe using a superconducting quantum intereference device (SQUID) magnetometer.[13] $^{57}$Fe-Mössbauer studies were recorded using a $^{57}$Co(Rh) source with a constant-acceleration spectrometer. Low-temperature spectra were taken in transmission geometry using a conventional bath cryostat as well as a cryostat equipped with a superconducting solenoid. The evaluation of the spectra with complex hyperfine interactions was performed with the RECOIL 1.03 fit routine.[14]

Two different absorbers were prepared from the single crystals of $Rb_{0.8}Fe_{1.6}Se_{2.0}$ for the Mössbauer studies. A powdered sample was prepared by crushing small pieces of the single crystals under strictly inert conditions in a glove box and compressing the powder between plastic windows of the absorber holder. This yields a textured sample with the crystalline $c$ axis oriented preferentially perpendicular to the absorber plane as evidenced from the $^{57}$Fe spectra. Another absorber was prepared by attaching thin single-crystalline flakes, separated from the bulk single crystals by the so-called "scotch-tape" technique. Mössbauer spectra of this sample reflect the quasi-single-crystalline quality of the absorber, called in the following the "mosaic" absorber, with the $c$ axis oriented perpendicular to the absorber plane and the $a,b$ axes randomly within the absorber plane, which resulted in a transmission of the gamma rays parallel to the $c$ axis within a range of $\pm 5°$.

Figure 1 exhibits the temperature dependence of zero-field-cooled (ZFC) and field-cooled (FC) susceptibilities of a single-crystalline $Rb_{0.8}Fe_{1.6}Se_{2.0}$ sample measured in a field of 10 Oe applied along the $c$ axis. The transition to a fully superconducting state occurs at $T_C = 32.4$ K within 1.5 K. The bulk nature of superconductivity was concluded from specific-heat measurements, yielding a volume fraction above 90%.[13] The inset in the Fig. 1 shows the magnetic hysteresis measured at 2 K after cooling in zero field. The width of
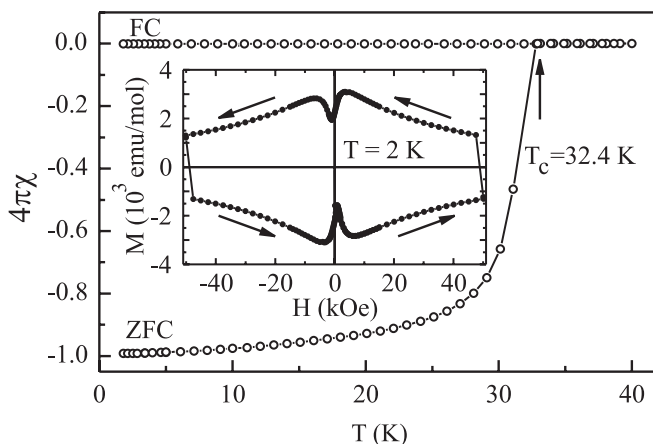
FIG. 2. $^{57}$Fe-Mössbauer spectra of $Rb_{0.8}Fe_{1.6}Se_2$ measured at $T = 4.2$ K of (a) a powdered sample and (b) a mosaic sample with the $c$ axis parallel to the direction of the gamma rays. (c) Mosaic sample as in (b) with an external magnetic field of 50 kOe applied parallel to the $c$ axis. Subspectra of the magnetic Fe sites are marked in dark gray, and subspectra of nonmagnetic Fe sites are shown in black. Insets in (a) and (b) indicate the quadrupole-split subspectra of the nonmagnetic Fe sites with different polarizations and in (c) the quadrupole-split nonmagnetic Fe sites experiencing the external field. The velocity scale is given with respect to $\alpha$-Fe at 295 K.

FIG. 1. Temperature dependence of the susceptibility of a single-crystalline $Rb_{0.8}Fe_{1.6}Se_2$ sample measured in zero-field-cooled (ZFC) and in magnetic field of 10 Oe applied along the $c$ axis (FC). The inset shows the magnetization hysteresis loop measured at $T = 2$ K.
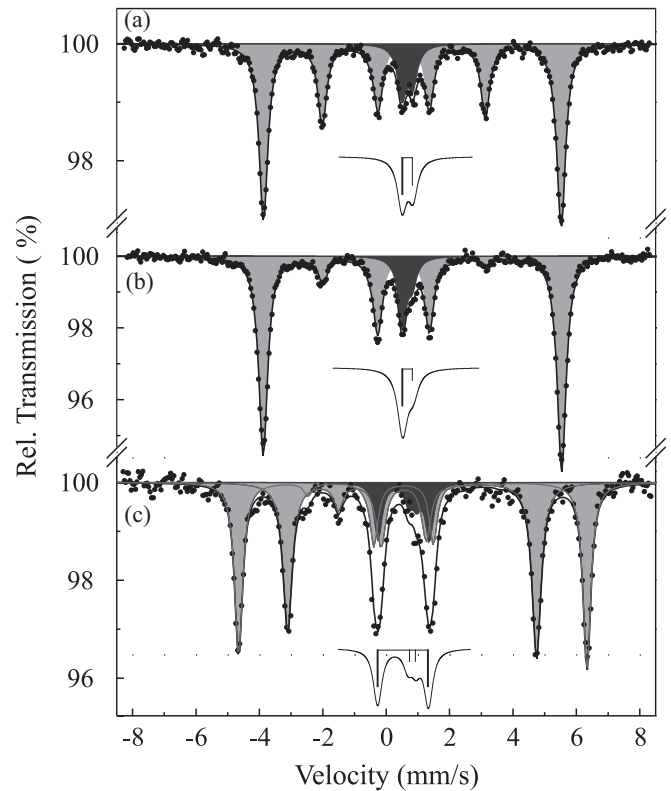
the diamagnetic response corresponds to a critical current density of $1.6 \times 10^4$ A/cm$^2$.[13] Finally, from resistivity studies, upper critical fields $H_{c2} = 25$ T with the magnetic field parallel to the $c$ axis and $H_{c2} = 63.3$ T for the perpendicular direction were derived.[13] These latter values are lower than the ones reported in other studies of $Rb_{1-x}Fe_{2-y}Se_2$, while the other superconducting properties compare well with those reported.[8,15]

Mössbauer spectra of the powdered sample were measured first at 4.2 K [Fig. 2(a)] and then with increasing temperature up to room temperature. All spectra are very similar to that observed at 4.2 K, as long as the absorber was kept under inert conditions. They consist of a dominant magnetic sextet with 88(1)% spectral intensity of the Fe sites and of a nonmagnetic quadrupole doublet with 12(1)% intensity. Both spectra show clear evidence for a strong texture by a deviation of the line intensity ratios of the magnetic subspectra from 3:2:1:1:2:3 and of the quadrupole doublet from a 1:1 ratio, as expected for a fully polycrystalline absorber. A fit analysis of the dominant magnetic subspectrum in Fig. 2(a) was only successful when a combined magnetic-dipole and electric-quadrupole interaction

was applied, where the direction of the magnetic hyperfine field does not coincide with the main axis of the electric-field gradient (EFG) tensor, including the asymmetry parameter $\eta$ for the EFG. As described in Refs. 9 and 10, the formation of the superstructure of $A_x\mathrm{Fe}_{2-y}\mathrm{Se}_2$ by an ordering of the vacant Fe sites is connected with a first-order structural phase transition, triggered by the concomitant onset of strong magnetic interactions within and between the supermoments formed by four Fe neighbors with parallel spins, coupled antiferromagnetically with the neighboring supermoments of Fe quartets. The point symmetry of the magnetic $16i$ sites is lowered by this structural phase transition from $4m$ to 1, as reflected by the changes of the Fe-Se and Fe-Fe distances as well as Se-Fe-Se binding angles, analyzed for the magnetic $\mathrm{K}_{0.8+x}\mathrm{Fe}_{1.6-y}\mathrm{Se}_2$ system.[16] The derived EFG tensor at the $16i$ sites, $\Delta E_Q = +1.23(1)$ mm/s with $\eta = 0.10(4)$ and an angle $\theta = 45(2)°$ between the main axis of the EFG and the magnetic hyperfine field $B_{\mathrm{hf}} = 283.0(3)$ kOe at 4.2 K, is drastically different in magnitude and direction to that in the undistorted Fe-Se layers of superconducting $\mathrm{Fe}_{1.01}\mathrm{Se}$ with $\Delta E_Q = -0.32$ mm/s at 4.2 K.[3,4] This large value of $\Delta E_Q$ can only originate from the local $\mathrm{Fe}^{2+}$ moment in a high-spin state, thereby providing a large orbital contribution to the EFG. From the relative intensity ratios of the magnetic six-line pattern, a preferred orientation of the magnetic hyperfine fields, and therewith of the crystalline $c$ axes, with an averaged angle of $40(2)°$ with respect to the direction of the $\gamma$ rays, is derived. The nonmagnetic site could be well adjusted by a quadrupole splitting with the same polarization dependence and $\Delta E_Q = -0.32(2)$ mm/s, which in sign and magnitude is identical to that observed in superconducting $\mathrm{Fe}_{1.01}\mathrm{Se}$ with the main axis of the EFG parallel to the $c$ axis.[3,4] This is important information showing that these nonmagnetic Fe sites are located either on the formally vacant $4d$ sites of the $\sqrt{5} \times \sqrt{5} \times 1$ superstructure of $\mathrm{Rb}_{0.8}\mathrm{Fe}_{1.6}\mathrm{Se}_2$ (Refs. 9 and 10) or in the above-mentioned nanosized phase with a $\sqrt{2} \times \sqrt{2} \times 1$ structure. It is important to note that the orientation of the $c$ axes of both phases coincide, as evidenced in the superstructure reflections presented for $\mathrm{K}_x\mathrm{Fe}_{2-y}\mathrm{Se}_2$.[11,12]

The spectrum of the mosaic sample shown in Fig. 2(b) provides direct information about the high degree of orientation of the $c$ axes of the magnetic phase and therewith of the magnetic hyperfine fields $B_{\mathrm{hf}}$ of the Fe $16i$ sites, evidenced by the strongly reduced intensities of lines 2 and 5. The magnetic subspectrum, again with 88% relative intensity, is well reproduced by the fit with the parameters given above for the powdered sample, providing also the unusual intensity ratios of the magnetic sextet, amounting to 2.86:0.30:1.04:0.96:0.10:3.14 [see Fig. 2(b), from the left to the right]. This indicates a pronounced deviation from the above-mentioned intensity ratio of purely magnetic sextets, which is caused by complex noncollinear hyperfine interactions and most pronounced for the present quasi-single-crystalline absorber, oriented with an averaged angle of $6(2)°$ between the direction of the $\gamma$ rays with respect to $B_{\mathrm{hf}}$ and, correspondingly, to the crystallographic $c$ axis. This information agrees well with the polarization dependence of the quadrupole doublet of the nonmagnetic sites, being now very close to 3:1, reflected by a fitted angle

of $8(3)°$ between the gamma rays and the main axis of the EFG, being therewith parallel to the $c$ axes of the mosaic sample.

Several $^{57}\mathrm{Fe}$-Mössbauer studies performed on $A_x\mathrm{Fe}_{2-y}\mathrm{Se}_2$ systems are known so far. $\mathrm{K}_{0.80}\mathrm{Fe}_{1.76}\mathrm{Se}_{2.00}$ was studied in the wide temperature range from 10 K up to and above the Néel temperature $T_N = 532$ K by Ryan et al.[17] $\mathrm{K}_{0.86}\mathrm{Fe}_{1.73}\mathrm{Se}_2$ was studied between 16 and 295 K by Li et al.[18] In both studies, the $^{57}\mathrm{Fe}$ spectra of mosaic absorbers prepared from single-crystalline flakes as in the present Rapid Communication, exhibited for the magnetic Fe sites the same spectral features as for the mosaic $\mathrm{Rb}_{0.8}\mathrm{Fe}_{1.6}\mathrm{Se}_2$ sample shown in Fig. 2(b), e.g., the same unusual intensity ratios of the dominant magnetic sextet pointing to the same spectral parameters for the Fe moments oriented parallel to the crystal $c$ axis. However, in both studies obviously a collinear quadrupole interaction was adjusted to the magnetic sites, quoted as $\Delta E_Q = +0.33(2)$ mm/s in Ref. 17 and with a similar value in Ref. 18, not quoted but extracted from an inspection of the fitted subspectra. By this fitting both studies could not account for the appearance of "anomalous" line intensities attributed to components 2 and 5 as well as for clearly visible asymmetries in line pairs 1–6 and 3–4. In addition, the line positions of the line pairs 2–5 and 3–4 could not be correctly adjusted. All these properties are well described in the present Rapid Communication with a higher statistical accuracy of the spectra than in Refs. 17 and 18. It is important that, for $\mathrm{K}_{0.80}\mathrm{Fe}_{1.76}\mathrm{Se}_{2.00}$, also a nonmagnetic component with a relative intensity of 12(2)% intensity, identical also in position to the present spectrum in Fig. 2(b), was observed,[17] however, the quadrupole splitting of this site was not analyzed because of the lower spectral resolution. The $^{57}\mathrm{Fe}$ spectrum of the mosaic absorber of $\mathrm{K}_{0.86}\mathrm{Fe}_{1.73}\mathrm{Se}_2$ (Ref. 18) exhibits a similar nonmagnetic component, but with a higher intensity of $\sim$25%, while a powdered absorber exhibits a nonmagnetic component of even higher intensity.[18] One can interpret this fact, at least in part, by decomposition products, similar to the observations in FeSe.[4] A recent Mössbauer study of powdered $A_{0.8}\mathrm{Fe}_{2-y}\mathrm{Se}_2$ samples, also of $\mathrm{Rb}_{0.8}\mathrm{Fe}_{2-y}\mathrm{Se}_2$,[19] provided similar spectra to the one shown in Fig. 2(a), however, due to the use of only powdered samples, with, in part, different conclusions, e.g., about the orientation of the Fe moments.

Using the advantage of a high degree of orientation of the crystalline $c$ axes of the mosaic $\mathrm{Rb}_{0.8}\mathrm{Fe}_{1.6}\mathrm{Se}_2$ sample, an additional $^{57}\mathrm{Fe}$ spectrum was measured at 4.2 K in an external magnetic field of $B_{\mathrm{ext}} = 50$ kOe applied parallel to the $c$ axes and to the direction of the gamma rays [Fig. 2(c)]. The spectral features are now drastically changed. The magnetic sites are split into two components of exact equal intensity with magnetic hyperfine fields of $B_{\mathrm{hf}}(1) = B_{\mathrm{hf}} + B_{\mathrm{ext}}$ and $B_{\mathrm{hf}}(2) = B_{\mathrm{hf}} - B_{\mathrm{ext}}$, with $B_{\mathrm{hf}} = -283.0(3)$ kOe. Therefore, $B_{\mathrm{hf}}(1)$ is attributed to Fe sites building up the supermoments with their combined moments arranged parallel to $B_{\mathrm{ext}}$, as demonstrated by the observed value $B_{\mathrm{hf}}(1) = -232.9(5)$ kOe, which is exactly the sum of $B_{\mathrm{hf}} + B_{\mathrm{ext}}$. Accordingly, $B_{\mathrm{hf}}(2) = B_{\mathrm{hf}} - B_{\mathrm{ext}} = -333.0(5)$ kOe is monitored by the Fe sites with their moments arranged antiparallel to $B_{\mathrm{ext}}$. The nonmagnetic Fe sites experience the same external field, and their quadrupole

spectrum is now split by $B_{ext}$ parallel to $V_{zz}$, the main component of the EFG, as shown in Fig. 2(c), with a fitted value of $B_{hf} = 50.1(5)$ kOe, exactly the value of $B_{ext}$. This is direct proof that the Fe sites in the paramagnetic phase do not possess a magnetic moment. The same observation was obtained from Mössbauer spectra of superconducting FeSe in an external field.[20]

Since the external field was applied parallel the $c$ axis of the oriented crystals of $Rb_{0.8}Fe_{1.6}Se_2$, the above observations prove the antiferromagnetic arrangement of the magnetic Fe sites in their fourfold supermoments arranged parallel or antiparallel to the $c$ axis, as derived from neutron-scattering studies for various $A_xFe_{1-y}Se_2$ systems.[9,21] The narrow linewidths of both magnetic and nonmagnetic Fe sites in all spectral features point to a rather perfect composition of the present $Rb_{0.8}Fe_{1.6}Se_2$ sample. Any kind of Fe off-stoichiometry at the $16i$ sites, e.g., only three Fe forming a ferromagnetic supermoment, should be reflected in the linewidth of the $16i$ sites or by additional magnetic sextets in the $^{57}Fe$ spectra with different hyperfine parameters, which is not the case. The present data disprove a possible arrangement with two different Fe moments, as discussed for the $Rb_{0.8}Fe_{1.6}Se_2$ system by Pomjakushin *et al.*[21] The latter case should be reflected by two different magnetic subspectra in Fig. 2(b) and four magnetic subspectra in Fig. 2(c), which are not observed.

One of the most important present finding is that 12% of the $Fe^{2+}$ ions are occupying a well-defined nonmagnetic site, following in its polarization dependence the principal orientation the $c$ axis of the $\sqrt{5} \times \sqrt{5} \times 1$ superstructure. The following discussion is devoted to attribute these nonmagnetic Fe sites either (i) to the formally empty $4d$ sites of the $\sqrt{5} \times \sqrt{5} \times 1$ superstructure or (ii) to the nonmagnetic minority phase, observed in $A_xFe_{1-y}Se_2$ superconductors as mentioned above.

(i) Considering an exact stoichiometry of the present sample as $Rb_{0.8}Fe_{1.6}Se_2$ and assuming a full occupancy of the $16i$ sites, as evidenced by the high spectral quality of the magnetic subspectra, there is no Fe left to occupy the "empty" $4d$ sites in the $\sqrt{5} \times \sqrt{5} \times 1$ superstructure. The actually observed 12% spectral intensity of nonmagnetic sites, assuming again a full occupancy of the $16i$ sites, would correspond to a $\sim$55% occupation of the $4d$ sites. This is a very high value which contradicts the so-called vacancy order of Fe $4d$ sites, which is considered to be a crucial factor for the formation of the magnetic superstructure.

From an XRD analysis of superconducting $K_{0.737}Fe_{1.613}Se_2$ and $K_{0.775}Fe_{1.631}Se_2$, occupancy of the $4d$ sites by 3.2% and by 7.8%, respectively, was reported, together with a full occupation of the $16i$ sites.[10] This contrasts with nonsuperconducting $K_{0.862(3)}Fe_{1.563(4)}Se_2$, where an $4d$-site occupancy as high as 22%, connected with 92% occupation of the $16i$ sites, was reported.[16] Summarizing, one can conclude that 55% occupation of the $4d$ sites is not compatible with an XRD analysis of superconducting $A_xFe_{2-y}Se_2$ systems as well as with the perfect magnetic properties of the $16i$ sites in the $\sqrt{5} \times \sqrt{5} \times 1$ superstructure evidenced in the present $^{57}Fe$ spectra.

(ii) Most important for the attribution of the nonmagnetic Fe sites to the nanosized minority phase is the fact that the principal $c$ axes of both majority and minority phases coincide. Now we can assign the observed 88% magnetic Fe sites in the $^{57}Fe$ spectra to the $16i$ sites of the $\sqrt{5} \times \sqrt{5} \times 1$ phase and the 12% nonmagnetic Fe sites to this minority phase, attributed in Refs. 11 and 12 to the superconducting properties of the whole sample. The identical direction of the $c$ axes of both phases explains immediately the observed polarization dependence of the quadrupole interaction of the nonmagnetic Fe sites, which is, in addition, identical in magnitude and polarization dependence to that observed in superconducting FeSe.

Finally we want to point out that it emerges from the numerous reports on the $A_xFe_{2-y}Se_2$ systems that a relative Fe amount of 1.60, within experimental uncertainty, marks the lower borderline for the occurrence of superconductivity. For instance, Wang *et al.*[22] reported that $Rb_{0.89}Fe_{1.58}Se_2$ is nonsuperconducting and exhibits only the $\sqrt{5} \times \sqrt{5} \times 1$ structure and strong magnetism with $T_N = 475$ K. For a nonsuperconducting $Rb_{0.73}Fe_{1.43}Se_2$ sample we observed magnetic $^{57}Fe$ spectra with identical hyperfine parameters to the present sample, but without nonmagnetic sites. All these findings support the location of the nonmagnetic sites into the nanoscaled minority phase. The present finding about a phase separation in $Rb_{0.8}Fe_{1.6}Se_2$ is supported by a following optical study of the same sample.[23] Also by NMR spectroscopy, evidence for a phase separation in $A_xFe_{2-y}Se_2$ systems is reported.[24]

Concluding, we have shown that $^{57}Fe$-Mössbauer spectra of powdered and single-crystalline samples of $Rb_{0.8}Fe_{1.6}Se_2$ exhibit well-resolved hyperfine spectra with a dominant magnetic site and a nonmagnetic site in an intensity ratio of 88(1)%–12(1)%. The magnetic sites can be perfectly adjusted by a fit analysis of $Fe^{2+}$ ions with noncollinear magnetic-dipole and electric-quadrupole interactions. The spectra of the single-crystalline absorber prove the orientation of the moments parallel to the crystalline $c$ axis. The spectra in an external magnetic field confirm the antiferromagnetic arrangement of ferromagnetically coupled Fe supermoments within the $\sqrt{5} \times \sqrt{5} \times 1$ superstructure.

The nonmagnetic Fe sites exhibit a quadrupole splitting with the same polarization dependence as the magnetic sites, indicating that they are located either on the formally vacant $4d$ sites of the dominant $\sqrt{5} \times \sqrt{5} \times 1$ superstructure or in a recently detected nanosized filamentary phase coexisting with the dominant magnetic phase. The present data favor the latter case, thereby supporting the findings from other $A_xFe_{2-y}Se_2$ systems that this nonmagnetic phase, with similar spectral properties as observed in superconducting FeSe, is also responsible for superconductivity in $Rb_{0.8}Fe_{1.6}Se_2$.

*v.ksenofontov@uni-mainz.de

[1] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. **130**, 3296 (2008); X. H. Chen, T. Wu, G. Wu, R. H. Liu, H. Chen, and D. F. Fang, Nature (London) **453**, 761 (2008); J. Zhao *et al.*, Nat. Mater. **7**, 953 (2008); M. Rotter, M. Tegel, and D. Johrendt, Phys. Rev. Lett. **101**, 107006 (2008); Z.-A. Ren *et al.*, Chin. Phys. Lett. **25**, 2215 (2008).

[2] F. C. Hsu *et al.*, Proc. Natl. Acad. Sci. USA **105**, 14262 (2008); Y. Mizuguchi, F. Tomioka, S. Tsuda, T. Yamaguchi, and Y. Takano, Appl. Phys. Lett. **93**, 152505 (2008); S. Margadonna, Y. Takabayashi, Y. Ohishi, Y. Mizuguchi, Y. Takano, T. Kagayama, T. Nakagawa, M. Takata, and K. Prassides, Phys. Rev. B **80**, 064506 (2009); V. Ksenofontov, G. Wortmann, A. I. Chumakov, T. Gasi, S. Medvedev, T. M. McQueen, R. J. Cava, and C. Felser, *ibid.* **81**, 184510 (2010).

[3] S. Medvedev, T. M. McQueen, I. Trojan, T. Palasyuk, M. I. Eremets, R. J. Cava, S. Naghavi, F. Casper, V. Ksenofontov, G. Wortmann, and C. Felser, Nat. Mater. **8**, 630 (2009).

[4] T. M. McQueen, Q. Huang, V. Ksenofontov, C. Felser, Q. Xu, H. Zandbergen, Y. S. Hor, J. Allred, A. J. Williams, D. Qu, J. Checkelsky, N. P. Ong, and R. J. Cava, Phys. Rev. B **79**, 014522 (2009).

[5] I. I. Mazin, Physics **4**, 26 (2011).

[6] F. Steglich, J. Aarts, C. D. Bredl, W. Lieke, D. Meschede, W. Franz, and H. Schäfer, Phys. Rev. Lett. **43**, 1892 (1979).

[7] J. Guo, S. Jin, G. Wang, S. Wang, K. Zhu, T. Zhou, M. He, and X. Chen, Phys. Rev. B **82**, 180520(R) (2010); A. Krzton-Maziopa, Z. Shermadini, E. Pomjakushina, V. Pomjakushin, M. Bendele, A. Amato, R. Khasanov, H. Luetkens, and K. Conder, J. Phys.: Condens. Matter **23**, 052203 (2011).

[8] A. F. Wang *et al.*, Phys. Rev. B **83**, 060512(R) (2011).

[9] Wei Bao, Q. Huang, G. F. Chen, M. A. Green, D. M. Wang, J. B. He, X. Q. Wang, and Y. Qiu, Chin. Phys. Lett. **28**, 086104 (2011).

[10] P. Zavalij *et al.*, Phys. Rev. B **83**, 132509 (2011); F. Ye, S. Chi, Wei Bao, X. F. Wang, J. J. Ying, X. H. Chen, H. D. Wang, C. H. Dong, and Minghu Fang, Phys. Rev. Lett. **107**, 137003 (2011);

V. Yu. Pomjakushin, D. V. Sheptyakov, E. V. Pomjakushina, A. Krzton-Maziopa, K. Conder, D. Chernyshov, V. Svitlyk, and Z. Shermadini, Phys. Rev. B **83**, 144410 (2011).

[11] R. H. Yuan, T. Dong, Y. J. Song, G. F. Chen, J. P. Hu, J. Q. Li, and N. L. Wang, e-print arXiv:1102.1381v3.

[12] A. Ricci *et al.*, Phys. Rev. B **84**, 060511(R) (2011); A. Ricci, N. Poccia, B. Joseph, G. Arrighetti *et al.*, Supercond. Sci. Technol. **24**, 082002 (2011).

[13] V. Tsurkan, J. Deisenhofer, A. Günther, H.-A. Krug von Nidda, S. Widmann, and A. Loidl, Phys. Rev. B **84**, 144520 (2011). The present sample is labeled BR16.

[14] K. Lagarec and D. G. Rancourt, Nucl. Instrum. Methods Phys. Res. B **129**, 266 (1997).

[15] C.-H. Li, B. Shen, F. Han, X. Zhu, and H.-H. Wen, Phys. Rev. B **83**, 184521 (2011).

[16] J. Basca, A. Y. Ganin, Y. Takabayashi, K. E. Christensen, K. Prassides, M. J. Rosseinsky, and J. B. Claridge, Chem. Sci. **2**(6), 1054 (2011).

[17] D. H. Ryan, W. N. Rowan-Weetaluktuk, J. M. Cadogan, R. Hu, W. E. Straszheim, S. L. Bud'ko, and P. C. Canfield, Phys. Rev. B **83**, 104526 (2011).

[18] Z. Li, X. Ma, H. Pang, and F. Li, e-print arXiv:1103.0098v3.

[19] I. Nowik, I. Felner, M. Zhang, A. F. Wang, and X. H. Chen, Supercond. Sci. Technol. **24**, 095015 (2011).

[20] A. Błachowski, K. Ruebenbauer, J. Żukrowski, J. Przewoźnik, K. Wojciechowski, and Z. M. Stadnik, J. Alloys Compd. **494**, 1 (2010).

[21] V. Yu. Pomjakushin, E. V. Pomjakushina, A. Krzton-Maziopa, K. Conder, and Z. Shermadini, J. Phys.: Condens. Matter **23**, 156003 (2011).

[22] M. Wang *et al.*, Phys. Rev. B **84**, 094504 (2011); e-print arXiv:1105.4675v1.

[23] A. Charnukha, J. Deisenhofer, D. Pröpper, M. Schmidt, Z. Wang, Y. Goncharov, A. N. Yaresko, V. Tsurkan, B. Keimer, A. Loidl, and A. V. Boris, e-print arXiv:1108.5698.

[24] L. Ma, G. F. Ji, J. Dai, J. B. He, D. M. Wang, G. F. Chen, B. Normand, and Weiqiang Yu, e-print arXiv:1103.4960v3.