Here we present additional SESANS and LD data on hematite (α-Fe$_2$O$_3$) and calcite (CaCO$_3$) powder samples. Besides the cylindrical vanadium (diameter 12 mm) and the flat quartz glass (width 2 mm) containers we used a flat aluminum container with an inner width of 2 mm and a window thickness of 0.3 mm, both in reflection and transmission geometry.

SESANS and LD experiments on CaCO$_3$ were conducted to get a further test for the LD data analysis procedure including SANS. The calcite powder for the LD experiment was previously well characterized in a high resolution synchrotron radiation (SR) diffraction study [1]. Calcite has a negligible contribution to incoherent neutron scattering.

### S1. SESANS

The results of the SESANS experiments are summarized in Table S1. The $\alpha_{\text{SESANS}}$ give the Gaussian full-width-at-half-maximum of the angular small angle distribution in degrees. The subscripts raw and norm indicate raw data and data normalized to the empty container, respectively.

The SANS of the empty containers is in the order of 0.05°, where the cylindrical vanadium container tends to be slightly better than aluminum and quartz glass. The $\alpha_{\text{SESANS}}$ increases with decreasing $k$ for both empty and filled containers. For the α-Fe$_2$O$_3$ sample, $\alpha_{\text{SESANS}}$ of the flat container is reduced by more than a factor 2 compared to the cylindrical container due to the smaller width of the latter.

The smaller $\alpha_{\text{SESANS}}$ for CaCO$_3$ results from the increased grain size of $\sim$ 400 nm (α-Fe$_2$O$_3$: $\sim$ 400 nm) and from a smaller scattering length density (2/3 of α-Fe$_2$O$_3$). The neutron absorption cross section of CaCO$_3$ is by a factor 14.5 smaller than for α-Fe$_2$O$_3$ and is thus negligible for our SESANS and LD experiments.

### S2. α − Fe$_2$O$_3$ (hematite)

Fig. S1 and Table S2 show additional LD data from α-Fe$_2$O$_3$ for the (116) and (214) reflections (for (024) see Fig. 7 in the main text). The contribution of SANS to the polarization decreases with increasing $k$, and is more pronounced for the cylindrical sample. After correction by the SESANS measured at $k = 1.7$ Å$^{-1}$ on the flat container (Fig. 5(a), main text), the agreement of the intrinsic polarization signal $P_{\text{inst}}$ for both container types is satisfactory, where the data from the flat container are more reliable due to the smaller correction. Both SR and LD data show significantly broader widths (Table S2) than those calculated from the monoclinic structure model established in ref. [2], although the widths follow the trend of the calculated values. The main contribution to the width is probably due to micro strains in the powder grains.

**TABLE S1. Summary of all SESANS experiments.**

| $k$ [Å$^{-1}]$ | sample | container | $\alpha_{\text{SESANS raw}}$ [°] | $\alpha_{\text{SESANS norm}}$ [°] |
|--------------|--------|-----------|-------------------------------|-------------------------------|
| 1.7          | empty  | flat quartz | 0.0618(7)                     | —                             |
| 1.7          | empty  | flat Al   | 0.0606(8)                     | —                             |
| 1.7          | α-Fe$_2$O$_3$ | flat quartz | 0.1278(17)                  | 0.1118(19)                  |
| 1.7          | α-Fe$_2$O$_3$ | flat Al   | 0.1208(18)                  | 0.1044(20)                  |
| 1.7          | CaCO$_3$ | flat Al   | 0.0705(9)                     | 0.0360(17)                  |
| 2.27         | empty  | flat Al   | 0.0560(21)                   | —                             |
| 2.27         | CaCO$_3$ | flat Al   | 0.0596(6)                    | 0.0203(18)                  |
| 2.27         | Fe$_2$O$_3$ | flat Al  | 0.0827(21)                   | 0.0541(32)                  |
| 2.3          | empty  | flat quartz | 0.0583(9)                   | —                             |
| 2.3          | α-Fe$_2$O$_3$ | flat quartz | 0.0878(6)                 | 0.0656(9)                   |
| 2.3          | empty  | cylinder | 0.0479(3)                    | —                             |
| 2.3          | α-Fe$_2$O$_3$ | cylinder | 0.1514(4)                   | 0.1439(6)                   |

**TABLE S2. Distribution of $d_{hkl}$ in α-Fe$_2$O$_3$ for the flat quartz container, the cylindrical vanadium container, from SR diffraction, and calculation using a monoclinic structure model. All values in this table are in units of 1 × 10$^{-4}$.$^\text{a}$**

| (hkl) | $d_{hkl}$ flat | $d_{hkl}$ cyl. | width SR | width calc. |
|-------|----------------|----------------|----------|-------------|
| (024) | 7.06(15)       | 7.10(40)      | 6.76(6)  | 2.22        |
| (116) | 6.32(15)       | 6.01(28)      | 6.19(5)  | 1.67        |
| (214) | 5.06(21)       | 4.91(30)      | 5.12(5)  | 0.95        |
| (300) | 5.53(21)       | —             | 5.18(5)  | 1.12        |

We also tested the alignment of the flat Al container in the transmission and reflection geometries, which means setting the flat sample surface perpendicular (reflection) or parallel (transmission) to $Q$. As the effective path length in the flat sample of width $t_0$ at the Bragg angle $\theta$ is $L_r = t_0/\sin(\theta)$ in reflection and $L_t = t_0/\cos(\theta)$ in transmission, the ratio $L_r/L_t = \tan(\theta)$ is unfavorable for the transmission geometry for $\theta > 45^\circ$. For the LD experiments we used $\theta = 55^\circ$, such that $L_t/L_r \approx 1.4$. This effective path length difference results in a larger Gaussian width of the LD raw data (without any SANS or other corrections). For the (116) reflection we get $\epsilon_{\text{raw}} = 9.41(5) \times 10^{-4}$ in transmission and 8.68(5) × 10$^{-4}$ in reflection.
FIG. S1. Comparison of the Larmor diffraction signal from α-Fe$_2$O$_3$ for the flat (closed symbols, solid lines) and cylindrical (open symbols, dotted lines) sample containers for the (116) and (214) reflections in α-Fe$_2$O$_3$, measured at $k = 2.26$ Å$^{-1}$ and $k = 2.57$ Å$^{-1}$, respectively.

S3. CaCO$_3$ (calcite)

Three Bragg peaks in CaCO$_3$ were studied by LD, where the scattering angle $2\theta = 110^\circ$ was kept constant, and $k$ was set to 1.68, 2.01, and 2.36 Å$^{-1}$ for the (113), (018), and (211) reflections, respectively. Fig. S2 shows the SESANS and LD polarization for these three peaks. As mentioned before, the SANS contributed significantly less to the LD polarization than in the case of α-Fe$_2$O$_3$. The comparison of the distribution widths with SR data from ID22 [1] is plotted in Fig. S3, the numbers are summarized in Table S3. The agreement of LD and SR data is less good than in the case of α-Fe$_2$O$_3$. There is no obvious reason for this discrepancy, both samples of CaCO$_3$ were taken from the same batch. Eventually environmental conditions such as humidity could have led to a change.

TABLE S3. Distribution of $d_{hkl}$ in CaCO$_3$. Comparison of LD, SR, and calculation. All values in this table are in units of $1 \times 10^{-4}$.

| (hkl) | $d_{hkl}$ | width SR | width calc. |
|-------|----------|----------|-------------|
| (113) | 5.78(7)  | 4.54(7)  | 1.64        |
| (018) | 6.20(15) | 5.08(7)  | 1.71        |
| (211) | 5.06(12) | 4.02(11) | 1.0         |

FIG. S2. Larmor diffraction data from CaCO$_3$ in a flat sample container (symbols) for the (113), (018), and (211) reflections, respectively.
FIG. S3. $\Delta d/d$-spacing distribution $\delta_{hkl}$ (FWHM) vs. $Q = 1/d_{hkl}$ of three Bragg peaks from CaCO$_3$, calcite, obtained from LD (flat container) and SR diffraction. The $d$-distribution widths $\delta_{hkl}$ are $5.80(6) \times 10^{-4}$, $6.23(9) \times 10^{-4}$, and $4.82(9) \times 10^{-4}$ for the (113), (018), and (211) reflections.

of the sample properties, as there was a time span of 9 years between the SR and the LD experiments. Both the SR and LD distribution widths are much broader than the width calculated from the monoclinic structure model established in ref. [1]. This means that the peak width is dominated by other mechanisms, such as micro strains. The variations of the peak widths, however, roughly follow the variations of the calculated values.

[1] R. Przenioslo, P. Fabrykiewicz and I. Sosnowska, Physica B 496, 49-56 (2016).

[2] R. Przenioslo, I. Sosnowska, M. Stękiel, D. Wardecki, and A. Fitch, Physica B, 449, 72-76 (2014).