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In situ investigation of the evolution of lattice strain and stresses in austenite and martensite during quenching and tempering of steel

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Abstract

Energy dispersive synchrotron X-ray diffraction was applied to investigate in situ the evolution of lattice strains and stresses in austenite and martensite during quenching and tempering of a soft martensitic stainless steel. In one experiment, lattice strains in austenite and martensite were measured *in situ* in the direction perpendicular to the sample surface during an austenitization, quenching and tempering cycle. In a second experiment, the $\sin^2\psi$ method was applied in situ during the austenite-to-martensite transformation to distinguish between macro- and phase specific micro-stresses and to follow the evolution of these stresses during transformation. Martensite formation evokes compressive stress in austenite that is balanced by tensile stress in martensite. Tempering to 748 K (475 °C) leads to partial relaxation of these stresses. Additionally, data reveals that (elastic) lattice strain in austenite is not hydrostatic but *hkl* dependent, which is ascribed to plastic deformation of this phase during martensite formation and is considered responsible for anomalous behavior of the 200_γ reflection.

Keywords: Martensite; Synchrotron X-ray diffraction (XRD); Residual stresses; Tempering; Steel

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1. Introduction

Modern steels are multi-phase materials. The response of a multi-phase material to an applied load is a function of the volume fraction, distribution, orientation and shape of the phases present [1,2], as well as of the presence and magnitude of internal stresses, which remain after processing [3]. On loading, the applied external forces are superimposed on the internal stresses [1,2].

Internal stresses can be classified by the length scale over which they equilibrate [1,4,5]. Macro-stresses (type I) act over large distances and are an average over all phases and grains present; micro-stresses vary from grain to grain and from phase to phase (type II) or within a single grain/phase (type III). Internal stress can arise as a consequence of inhomogeneous elastic and thermal properties [2,3], inhomogeneous plastic strain [2], or a phase transition occurring in association with a shape change [5].

The austenite (γ)-to-martensite (α') transformation in steel is associated with a shape change, the so called transformation strain, which consists of a volume expansion of approx. 3% and a shear [6-9]. Additionally, austenite and martensite have different thermal and elastic properties. Thus, martensite formation is associated with the development of residual stress in the material, with contributions from the transformation itself as well as thermal mismatch [3,9].

The development of macro-stresses during quenching of steel parts is a well described subject [3-5,9,10]. Similarly, the generation of lattice defects (micro-stresses of type III) in austenite during martensite formation has been investigated in details [6-8]. On the other hand, the evolution of micro-stresses of type II is controversial: martensite formation has been reported to invoke compressive stress [11-22], tensile stress [22,23], or no stress [24-27] in austenite, while information about the stress state in the developing martensite is incomplete. The evolution of micro-stresses of type II during martensite formation is of fundamental interest because these stresses affect the transformation kinetics [28].

In the absence of macro-stresses (type I), the grain- or phase-specific micro-stresses (type II) can be evaluated from the lattice strain as experimentally determined by X-ray diffraction (XRD) [1,4] by measuring in the direction i the lattice spacing, $d_i^{hkl\varphi}$, for a given family of lattice planes, $\{hkl\}$, in the crystalline phase φ . Comparison of the measured lattice spacing with a reference lattice spacing, $d_{ref}^{hkl\varphi}$, provides the lattice strain, $\varepsilon_i^{hkl\varphi}$:

$$\varepsilon_i^{hkl\varphi} = \frac{d_i^{hkl\varphi} - d_{ref}^{hkl\varphi}}{d_{ref}^{hkl\varphi}} \quad (1)$$

55 The hkl -specific strain $\varepsilon_i^{hkl\varphi}$ represents the average lattice strain, $\langle \varepsilon^{hkl\varphi} \rangle$, for a given
56 family of lattice planes $\{hkl\}$ in the probed volume and can be converted into an average
57 (hydrostatic) stress, $\langle \sigma^\varphi \rangle$, applying the appropriate X-ray elastic constants, XECs [4]:

$$58 \quad \langle \varepsilon^{hkl\varphi} \rangle = \left(3 \cdot S_1^{hkl\varphi} + \frac{1}{2} S_2^{hkl\varphi} \right) \cdot \langle \sigma^\varphi \rangle \quad (2)$$

59 where $S_1^{hkl\varphi}$ and $\frac{1}{2} S_2^{hkl\varphi}$ are the XECs of phase φ for probing the family of lattice planes $\{hkl\}$.

60 Experimental XRD work on the evolution of lattice strain and phase-specific stresses in steel during
61 martensite formation was firstly reported in 1957 [11]. It was suggested that a low content of
62 retained austenite is in a state of compressive stress. In the same year, comparison of the lattice
63 parameter of Fe-30%Ni austenite in as-received condition and after approx. 80 % transformation by
64 sub-zero Celsius treatment did not show a significant change [24]. Therefore, it was concluded that
65 martensite formation did not evoke micro-stresses of type II in austenite.

66 More than 10 years later [12], Ridley et al. reproduced the results in Ref. [11] and suggested that
67 data could be interpreted in terms of a state of compression in austenite caused by martensite
68 formation. Alternatively, data could indicate depletion of austenite in C during quenching and room
69 temperature storage of the material. Nevertheless, in a study published 1 year earlier, Yeshov and
70 Oslon followed a^γ during quenching of several steel samples by probing $\{200\}_\gamma$ lattice planes and
71 showed the build-up of a tensile state of stress in austenite at the beginning of the transformation,
72 followed by relaxation of these stresses at a later stage [23]. Remarkably, later work by the same
73 authors [22] showed that martensite formation evokes a state of tensile lattice strain in austenite for
74 probing $\{200\}_\gamma$, while probing $\{111\}_\gamma$ showed compression. These results seem irreconcilable.

75 However, it should be realized that for f.c.c. metals lattice strains depend non-linearly on the
76 applied load if strained into the plastic region [29-32]. This non-linear behavior is hkl dependent
77 and yields anisotropic strains that are retained upon unloading. The $[200]_\gamma$ direction is elastically
78 the most compliant and plastically the softest and therefore has the most pronounced non-linear
79 behavior. Consequently, the 200_γ reflection is inappropriate for the evaluation of the average state
80 of residual stress in f.c.c. metals subjected to plastic deformation. According to Refs. [29-31],
81 probing 311_γ , or averaging lattice strains over several reflections, is most appropriate. Later,
82 reconsidering the results in Ref. [30], it was suggested that 111_γ or 422_γ are the most appropriate
83 reflections [32]. Evidently, an incorrect choice of the reflections probed for the evaluation of the
84 state of stress in austenite may have played a role in the discrepancy between data.

85 The first serious attempt to solve the controversy appeared in 1974, when Golovchiner determined
86 *in situ* the evolution of lattice strain in austenite versus the fraction transformed in a large number of

87 ferrous alloys [13]. These alloys were fully austenitic at room temperature and largely transformed
88 to martensite during cooling to 93 K (-180 °C). Determination of a^γ was performed based on the
89 311_γ reflection. In all investigated samples, compression was observed in austenite after martensite
90 formation for transformed fractions beyond 10-60 %, depending on the chemical composition of the
91 alloy.

92 In 1980, the focus firstly included the state of stress in martensite [14]. It was claimed that
93 martensite formation evoked compressive stresses in both austenite and martensite. Nevertheless,
94 this claim violates the condition that, in absence of external forces, balancing of forces should be
95 obtained for a finite matrix [3-5]:

$$96 \quad \sum_{\varphi} f^{\varphi} \cdot \langle \sigma^{\varphi} \rangle = 0 \quad (3)$$

97 with f^{φ} volume fraction of phase φ . Hence:

$$98 \quad f^{\alpha'} \cdot \langle \sigma^{\alpha'} \rangle = -f^{\gamma} \cdot \langle \sigma^{\gamma} \rangle \quad (4)$$

99 with $f^{\alpha'}$ and f^{γ} the volume fraction of martensite and austenite, respectively. Thus, compression in
100 austenite should be balanced by tension in martensite. It is important to realize that the lattice
101 strains determined in Ref. [14], departed from the assumption that stresses were null at the
102 beginning of the transformation. This is unlikely for the minority phase, martensite (cf. Eq. 4),
103 suggesting that the analysis may have been faulty. Additionally, data may have been affected by the
104 presence of macro-stresses, implying that Eq. 2 could not be used. Unfortunately, similar arguments
105 apply for all references cited above.

106 The need for more thorough stress analysis was firstly recognized in 1990 [15]. XRD was applied at
107 room temperature to determine a_{γ} (based on 220_γ) in Fe-C and Fe-N thin foils. Data indicated that
108 retained austenite experienced compressive strain in the direction perpendicular to the sample
109 surface. To convert strain into stress, measurements were performed in various directions. Because
110 of the shallow penetration depth of laboratory X-rays, all components of stress in martensite in the
111 direction perpendicular to the sample surface, $\sigma_{\perp}^{\alpha'}$, were assumed equal to zero. The principal stress
112 component parallel to the sample surface, $\sigma_{\parallel}^{\alpha'}$ was evaluated applying the $\sin^2\psi$ method (cf. Ref.
113 [4]). Data revealed that stresses in martensite were negligible within the probed volume. Since a
114 negligible state of stress in the majority phase (i.e. martensite) does not imply that the balancing
115 stress in the minority phase (i.e. austenite) is negligible, (cf. Eq.4) compressive strain in austenite
116 was interpreted in terms of a state of compressive stress in this phase.

117 The limitations associated with the shallow penetration/information depth of laboratory X-rays
118 could be overcome by applying Neutron diffraction [25,26] or synchrotron XRD [16-20,27].

119 In Refs. [25,26], a^γ was measured in homogeneous Fe-Ni-C austenite as well as after partial
120 transformation of the austenite into martensite. Data indicated that martensite formation did not
121 affect a^γ . Similarly, San. Martin et al. followed the formation of martensite in Fe-Cr-Ni-Mo
122 maraging steel up to 40 % fraction transformed and showed that the averaged value of a^γ does not
123 change with $f^{\alpha'}$ [27]. Conversely, Villa et al. showed that martensite formation during sub-zero
124 Celsius treatment of steel induces compressive lattice strain in austenite [16-18]. The application of
125 the $\sin^2\psi$ method ex situ indicated that lattice strain was caused by compressive stress in austenite,
126 which was hydrostatic within the probed volume. Similarly, Epp showed that compressive
127 hydrostatic stress build up in austenite during quenching of high C steel [19,20]. Evidently,
128 diffraction experiments at large scale facilities did not provide a unanimous answer.

129 A last attempt to clarify the picture was presented in Ref. [21]. Laboratory XRD was applied to
130 measure a^γ before and after martensite formation in a Fe-Ni alloy and data was complemented with
131 measurement of a^γ after tempering to 523 K (250°C). Tempering is expected, at least partially, to
132 lead to relaxation of internal stresses. Data showed that compressive lattice strain develops in
133 austenite during martensite formation for $f^{\alpha'} > 75\%$, and that this strain is largely annihilated during
134 tempering. No strain was observed in martensite. Compressive strain in austenite was interpreted in
135 terms of hydrostatic compressive stress in this phase.

136 Summarizing, no consistent picture exists of the evolution of stress in austenite and martensite
137 during martensite formation in steel. Careful evaluation of the discussed literature reveals that
138 during martensite formation a state of compressive stress builds-up in austenite. The compression is
139 observed by XRD only after a threshold fraction of austenite has transformed to martensite and
140 appears hydrostatic when averaged over the probed volume. The internal stress in austenite is
141 interpreted as micro-stress of type II, but this hypothesis has never been verified with the
142 simultaneous observation of balancing tensile micro-stress of type II in martensite. Tempering may
143 promote relaxation of stresses and give indication on their actual presence. In the present work,
144 experiments were designed to investigate the evolution of phase specific stresses in both austenite
145 and martensite during quenching and tempering of steel.

146 **2. Material and methods**

147 The material selected for investigation was commercial EN 1.4418 steel (Table 1), which is a soft
148 martensitic stainless steel grade, with a low fraction of interstitials. Martensite in this alloy is body
149 centered cubic b.c.c. and has lath morphology [33,34]. The martensite start temperature, M_s , equals
150 408 K (135 °C) [34] and the Curie temperature, T_C , of martensite equals 898 K (625 °C) [34].

151 Two Ø10 mm x 0.15 mm disks, labelled sample 1 and sample 2, respectively, were prepared for
152 synchrotron XRD investigation. Preparation comprised austenitization at 1223 K (950 °C) for 10
153 min in a continuous flow of Ar gas, followed by cooling to room temperature at an average rate of
154 approx. 1 K s⁻¹. Thereafter, samples were ground and electro-polished for 5 s at 40 V in Struers A-2
155 electrolyte. The condition of the material after preparation is labelled “as normalized”.

156 Energy-dispersive XRD investigation was performed in situ at the EDDI beamline of the BESSY II
157 synchrotron facility [35]. In Energy dispersive XRD, the investigation is carried out using a beam of
158 white X-rays at a fixed diffraction angle, 2θ , and diffraction for crystallographic planes, $\{hkl\}$, will
159 occur at a photon energy, E^{hkl} . The energy position, E^{hkl} , of the diffraction lines in the energy
160 spectrum is inversely proportional to the interplanar spacing, d^{hkl} [36]:

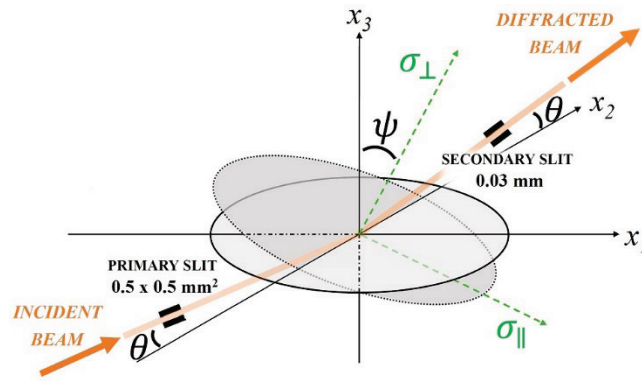
$$161 \quad E^{hkl} = \frac{hc}{2 \sin \theta} \cdot \frac{1}{d^{hkl}} \quad (5)$$

162 with h Planck’s constant and c the speed of light. The average information depth for each
163 diffraction line is a function of its energy, and of the diffraction geometry applied [37].

164 In the present investigation, the acquisition time was set to 60 s per spectrogram (i.e. diffractogram)
165 and the experiments were conducted with a 0.5 x 0.5 mm² slit configuration on the primary side
166 applying the diffraction geometry $\theta - 2\theta = 7^\circ - 14^\circ$. The equatorial slit aperture was set to 0.03
167 mm. Two configurations were applied: in the first, the scattering vector was kept perpendicular to
168 the sample surface; in the second, the angle between the scattering vector and the surface normal, ψ ,
169 was varied by tilting the sample around an axis parallel to the plane through incident and diffracted
170 beam ($\sin^2\psi$ method). The experimental setup is presented in Fig. 1.

171 Diffraction lines were fitted with pseudo-Voigt profile functions. Each reflection was analyzed
172 independently to account for anisotropic lattice strain. A gold reference standard was used for
173 calibration of the absolute value of the lattice parameter. The quantification of f^γ and $f^{\alpha'}$ was
174 based on a direct comparison of the integrated intensity of the diffraction peaks of austenite and
175 martensite [38]. Additional details were given in Ref. [33]. The analysis included the 200_γ , 220_γ ,
176 311_γ and 222_γ reflections of austenite and the $200_{\alpha'}$, $211_{\alpha'}$, $220_{\alpha'}$ and $321_{\alpha'}$ reflections of
177 martensite. The $111_\gamma/110_{\alpha'}$ couple was excluded because these reflections largely overlap and
178 because these line profiles correspond to the lowest information depth among all reflections and,
179 thus, are most sensitive for surface effects.

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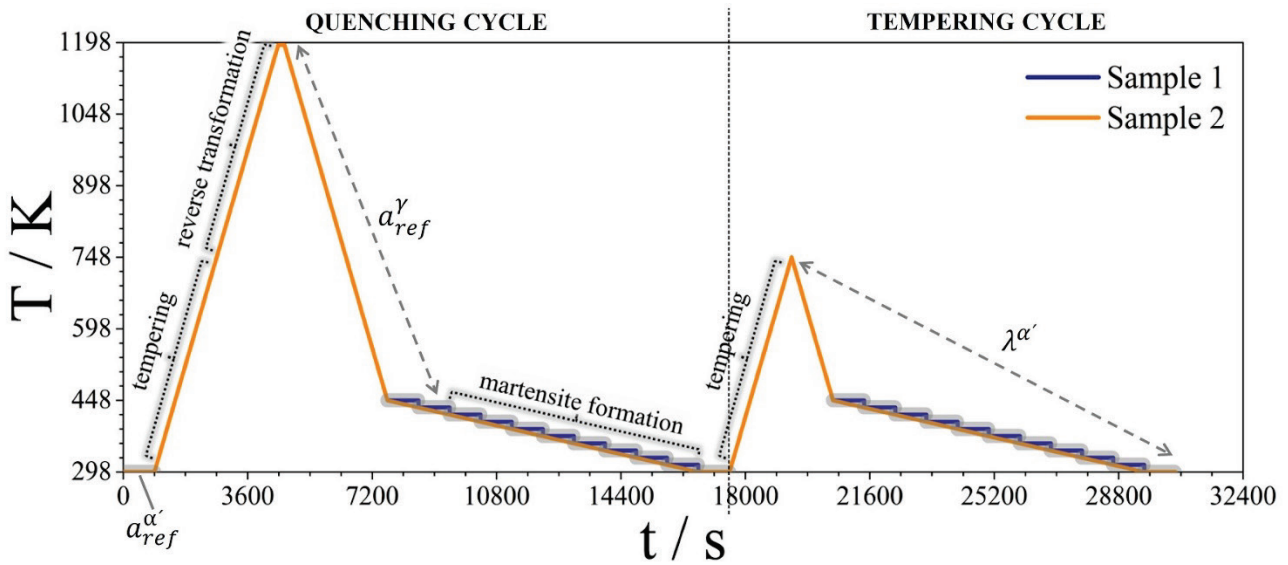
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Figure 1. Illustration of the applied experimental setup. The scattering vector is parallel to x_3 . The tilting axis is x_2 . ψ is the angle between the surface normal and the scattering vector.



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Figure 2. Schematic representation of the thermal cycle applied during the XRD investigation and of the various transformations occurring in the material. The graph also indicates the data which were used to estimate the reference values of the lattice parameters of austenite and martensite (see section 3.1.).

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Investigation was performed in an Anton Paar DHS 1100 Domed Hot Stage under continuous flow of protective Ar gas at a constant pressure of 1.4 bar. The samples were placed on the heating element (alumina plate) onto which they were secured by mild clamping. The temperature was measured with a Pt-Pt10Rh thermocouple fixed to the stage.

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The thermal cycle consisted of a quenching step, immediately followed by a tempering step and it is illustrated in Fig. 2. During the quenching step, samples were heated to 1193 K (920 °C) at a rate of 0.25 K s⁻¹, austenitized for 180 s, and cooled to 448 K (175 °C) at the same rate. Cooling from 448 K (175 °C) to room temperature was performed at an average rate of 0.017 K s⁻¹. The tempering step consisted of heating the samples to 748 K (475 °C) at a rate of 0.25 K s⁻¹ followed by continuous cooling under the same experimental conditions applied during quenching. The cooling steps from 448 K (175 °C) to room temperature were carried out as follows: sample 1 was cooled in

201 steps of 15 K maintaining an overall average cooling rate of 0.017 K s^{-1} and ψ , was varied from 0°
202 to 72° in steps of 8° at each investigation temperature (in situ $\sin^2\psi$ method); sample 2 was
203 continuously cooled at a rate of 0.017 K s^{-1} and ψ was maintained equal to 0° . Additionally, the
204 $\sin^2\psi$ method was applied ex situ at the beginning of the thermal cycle and at the end of the
205 investigation.

206 **3. Results and interpretation**

207 **3.1. Evaluation of lattice strains and stress analysis**

208 The calculation of lattice strains in austenite, $\varepsilon_{\psi}^{hkl\gamma}$, and martensite, $\varepsilon_{\psi}^{hkl\alpha'}$, from Eq.1 requires that
209 the strain-free lattice parameter of austenite, a_{ref}^{γ} , and of martensite, $a_{ref}^{\alpha'}$, are known as a function
210 of temperature.

211 The value of a_{ref}^{γ} was obtained by extrapolating a second order polynomial fitted through a_{\perp}^{γ}
212 (averaged over all hkl) as measured during cooling in the temperature interval 418-1193 K (145-920
213 $^\circ\text{C}$), where austenite is the only phase present because $T > M_s$. In particular, $\lambda^{\gamma} = 1.285 \cdot 10^{-5} +$
214 $1.052 \cdot 10^{-8} T - 4.677 \cdot 10^{-12} T^2$, with T temperature expressed in K, which is consistent with
215 literature data for austenitic stainless steels in the same temperature interval [45].

216 A similar experimental condition for $a_{ref}^{\alpha'}$, where martensite is the only phase present in the sample,
217 is not available. Therefore, the value of $a_{ref}^{\alpha'}$ at 298 K (25 $^\circ\text{C}$) was calculated under the condition
218 of balancing micro-stresses of type II (cf. Eq.4) at the beginning of investigation. The value of $a_{ref}^{\alpha'}$
219 for the temperature interval 298-898 K (25-625 $^\circ\text{C}$) was calculated from $a_{ref}^{\alpha'}$ at 298 K (25 $^\circ\text{C}$) and
220 the coefficient of thermal expansion of strain-free martensite, $\lambda^{\alpha'} = 1.266 \cdot 10^{-5} \text{ K}^{-1}$, as evaluated
221 by fitting $a_{\perp}^{\alpha'}$ as measured during cooling tempered martensite from 748 K (475 $^\circ\text{C}$) to room
222 temperature. This is consistent with literature data for martensitic stainless steel in the same
223 temperature interval [45]. For $T > 898 \text{ K}$ (625 $^\circ\text{C}$), martensite is paramagnetic and $\lambda^{\alpha'}$ evaluated for
224 ferromagnetic martensite does no longer apply [45]. This is beyond the scope of the present work.

225 Lattice strains were converted into stresses. During all thermal steps where $\psi = 0$, the average
226 stresses in the phases, $\langle \sigma^{\varphi} \rangle$, could be determined only under the assumption that the samples are
227 free from macro-stresses (cf. Eq. 2). Unfortunately, this assumption does not apply in the present
228 case, as will be shown in section 3.3.

229 In a thin sample, macro-stresses can be considered relaxed, and thus equal to zero, in the direction
230 normal to the sample surface, \perp . In the direction parallel to the sample surface, \parallel , macro-stresses

231 balance over the sample cross section, but are not necessarily balanced within the volume of
 232 material probed by XRD, and can thus deviate from zero.

233 To account for the possible presence of macro-stresses, lattice strains were measured in sample 1 at
 234 several ψ angles at fixed temperature. This procedure was applied only to the part of the thermal
 235 cycle of most interest for the current investigation. Stresses were determined under the assumption
 236 of rotational symmetry within the plane of the sample and the stress components $\sigma_{\parallel}^{\varphi} - \sigma_{\perp}^{\varphi}$ and σ_{\perp}^{φ}
 237 were calculated applying the $\sin^2\psi$ method. Assuming that the stress is rotationally symmetric, the
 238 lattice strain is given by [4]:

$$239 \quad \varepsilon_{\psi}^{hkl\varphi} = S_1^{hkl\varphi} [2 \cdot \sigma_{\parallel}^{\varphi} + \sigma_{\perp}^{\varphi}] + \frac{1}{2} S_2^{hkl\varphi} \cdot \sigma_{\perp}^{\varphi} + \frac{1}{2} S_2^{hkl\varphi} [\sigma_{\parallel}^{\varphi} - \sigma_{\perp}^{\varphi}] \cdot \sin^2\psi \quad (6)$$

240 The XEC $S_1^{hkl\varphi}$ and $\frac{1}{2} S_2^{hkl\varphi}$ (Table 2) were calculated from the single crystal elastic constants for
 241 ferrite [39] and austenite [40][†] applying the Eshelby/Kröner model for elastic grain interaction
 242 [41,42][‡] and the material is assumed free of texture.

243 For Energy Dispersive XRD, and under the assumption that macro-stresses in the \perp direction are
 244 null, the slope of the dependence of lattice strain, $\varepsilon_{\psi}^{hkl\varphi}$, on $\sin^2\psi$ provides $\sigma_{\parallel}^{\varphi} - \sigma_{\perp}^{\varphi}$ while the
 245 intercept equals $S_1^{hkl\varphi} [2 \cdot \sigma_{\parallel}^{\varphi} + \sigma_{\perp}^{\varphi}] + \frac{1}{2} S_2^{hkl\varphi} \cdot \sigma_{\perp}^{\varphi}$. The equations for slope and intercept provide
 246 the set of equations to assess the individual values of $\sigma_{\parallel}^{\varphi}$ and σ_{\perp}^{φ} , provided that the XECs are
 247 known. The macro-stress $\sigma_{\parallel}^{\varphi} - \sigma_{\perp}^{\varphi}$ (of type I) in phase φ corresponds to an hkl -dependent depth,
 248 ζ^{hkl} , and σ_{\perp}^{φ} represents the average micro-stresses of type II in φ . From Eq.3 it is obtained:

$$249 \quad \sum_{\phi} f^{\varphi} \cdot \langle \sigma_{\perp}^{\varphi} \rangle = 0 \quad (7)$$

250 where $\langle \sigma_{\perp}^{\varphi} \rangle$ represents the stress averaged over all investigated hkl for phase φ .[§]

251 In the present case, ζ^{hkl} can be estimated using the absorption coefficient of Fe for the appropriate
 252 energy and equals to approx. 4 μm , 5 μm , 14 μm , 22 μm and 25 μm for the 111 $_{\gamma}$, 200 $_{\gamma}$, 220 $_{\gamma}$,
 253 311 $_{\gamma}$ and 222 $_{\gamma}$ reflections of austenite, respectively, and approx. 4 μm , 10 μm , 18 μm , 27 μm and
 254 55 μm for the 110 $_{\alpha'}$, 200 $_{\alpha'}$, 211 $_{\alpha'}$, 220 $_{\alpha'}$ and 321 $_{\alpha'}$ reflections of martensite, respectively.

[†] The bulk elastic modulus of austenite, B^{γ} , and of martensite, B^{α} calculated from Refs. [51,52] are 184 GPa and 167 GPa, respectively. From Ref. [56], for Fe-15%Cr-5%Ni $B^{\gamma} = 164$ GPa and is not significantly affected by the presence of interstitials.[57] This indicates that Ref. [52] most likely overestimated the stiffness of austenite. No accurate information is available to evaluate the value of B^{α} from Ref. [51].

[‡] It should be noted that the Eshelby/Kröner model for the present case is an approximation, as it assumes elastic interaction of crystals with identical elastic constants, while the present material is two phase with different elastic constants for the two phases.

[§] The multiplicity of the various hkl was not taken into account, implying that all reflections was equally weighted.

255 3.2. Evolution of phase fraction and lattice strain during thermal cycling

256 3.2.1. Phase fraction

257 The evolution of the fraction of martensite as measured for diffraction vector perpendicular to the
258 surface, $f_{\perp}^{\alpha'}$, is shown in Fig. 3a versus temperature, T. Data plotted in Fig. 3a represents the
259 average value over the two investigated samples and is only representative of the fraction of
260 martensite in the material, $f^{\alpha'}$, for the (unlikely) assumption of random grain orientation [46].

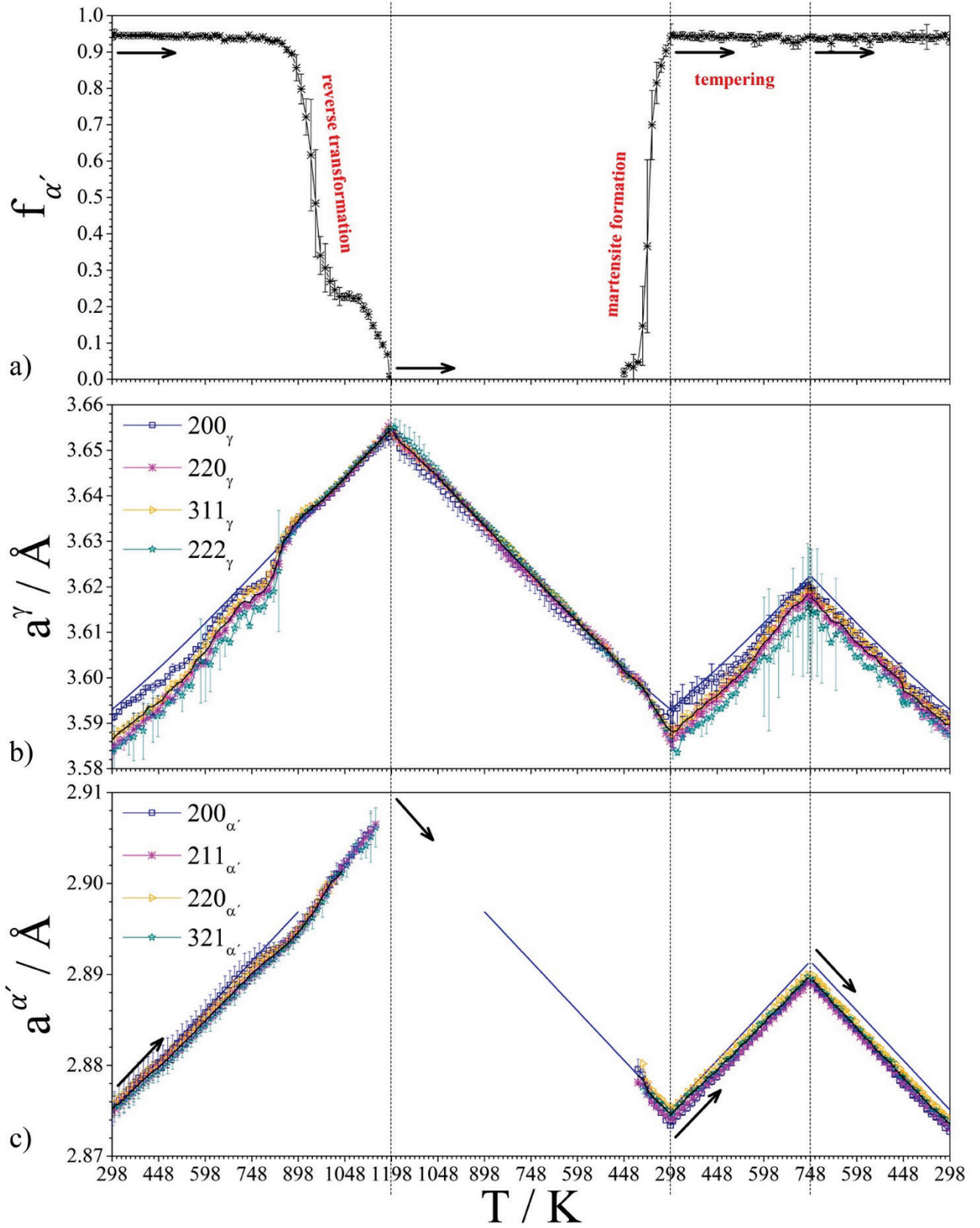
261 At the beginning of the investigation $f_{\perp}^{\alpha'}$ is 0.93 (Fig. 3a). During heating, reverse austenite
262 formation starts at about 823-848 K (550-575 °C). Austenitization proceeds in two steps and is
263 completed just below 1193 K (920 °C). Two-step kinetics of reverse austenite formation in soft
264 martensitic stainless steel is discussed in detail elsewhere [33,34,47]

265 On cooling from the austenitization temperature, martensite is firstly observed at 703 K (330 °C),
266 approx. 200 K above M_S , by appearance of the $110_{\alpha'}$ line profile, which is probed at the lowest
267 average information depth of all hkl s. On continued cooling, the intensity of $110_{\alpha'}$ increases and is
268 accompanied by $200_{\alpha'}$, $211_{\alpha'}$, $220_{\alpha'}$ and $321_{\alpha'}$ in the order of mentioning, consistent with a
269 gradual increase of the information depth ζ^{hkl} . Evidently, martensite formation starts at 703 K (330
270 °C) in the near surface region and progresses in the depth direction on continued cooling. It has
271 repeatedly been reported that the formation of lath martensite starts at the free surface at a
272 temperature significantly higher than M_S [47-52]. The present results are fully consistent with these
273 observations.

274 The martensite content $f_{\perp}^{\alpha'}$ exceeded 0.03 at about 403 K (130 °C), which is below M_S . Thereafter,
275 on continued cooling, a large fraction of martensite formed within the temperature range 383-353 K
276 (115-85 °C) followed by martensite formation at a progressively reduced rate. At 298 K (25 °C),
277 about 93 % of the austenite had transformed into martensite, consistent with the starting condition.

278 Upon quenching, the material was tempered to 748 K (475 °C) without affecting the fraction of
279 martensite in the sample.

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Figure 3. a) Fraction of martensite evaluated in the direction normal to the sample surface $f_{\perp}^{\alpha'}$ plotted versus temperature T (values represent average values for sample 1 and sample 2; the error bars are the standard deviation); b) lattice parameter of austenite measured in the direction normal to the sample surface a_{\perp}^{γ} plotted versus temperature T ; c) lattice parameter of martensite measured in the direction normal to the sample surface $a_{\perp}^{\alpha'}$ plotted versus temperature T . The blue continuous lines in b) and c) represent the strain free lattice parameter of the phases a_{ref}^{ϕ} . The black continuous lines in b) and c) show the value of the lattice parameter of the phases a_{ref}^{ϕ} averaged over the probed reflections, not including $200_{\alpha'}$. In order to compare between the two data sets, 1 data point every 15 K is considered for sample 2 when cooled within the temperature range $298 \text{ K} < T < 448 \text{ K}$ ($25 \text{ }^{\circ}\text{C} < T < 175 \text{ }^{\circ}\text{C}$).

290 3.2.2. *Lattice strains*

291 The lattice parameters calculated from the lattice spacing for $\{hkl\}$ are given for austenite in Fig. 3b
292 and for martensite in Fig. 3c. The evolutions of lattice strains in these phases are described
293 separately below.

294 *Austenite*

295 At the beginning of the quenching cycle, a^γ was significantly smaller than a_{ref}^γ (given by the
296 drawn blue line) and varies with the hkl probed, which indicates that austenite experiences
297 anisotropic compressive lattice strain in the direction \perp .

298 On heating, a^γ expands at a (hkl -dependent) rate which deviates from the value expected from the
299 thermal expansion coefficient λ_γ , indicating changes in lattice strain, $\varepsilon_\perp^{hkl\gamma}$. Compressive lattice
300 strains $\varepsilon_\perp^{hkl\gamma}$ increase up to 523 K (250 °C), decrease in the temperature range 523-748 K (250-475
301 °C), where after they increase again up to 798 K (525 °C), and eventually are annihilated at about
302 848 K (575 °C), when the reverse austenite formation commences.

303 On cooling, $a^\gamma = a_{ref}^\gamma$ for $T \geq 418$ K (145 °C) (by definition), implying that $\varepsilon_\perp^{hkl\gamma} = 0$. Thereafter,
304 for temperatures below M_s , $a^\gamma < a_{ref}^\gamma$, which indicates that martensite formation is accompanied
305 by compressive lattice strain in austenite in the direction perpendicular to the surface. The observed
306 lattice strain is largest at 298 K (25 °C) and depends on hkl : the largest $\varepsilon_\perp^{hkl\gamma}$ is observed for 220_γ
307 and 222_γ , while $\varepsilon_\perp^{hkl\gamma}$ is negligible for 200_γ and for 311_γ it shows a value close to the average
308 over hkl . This behaviour for the various hkl is analogous to the dependence on hkl for the starting
309 condition.

310 On tempering, the evolution of $\varepsilon_\perp^{hkl\gamma}$ during heating to 748 K (475 °C) reflects the same trends as
311 on heating the “as normalized” condition. On reaching 748 K (475 °C), about half the compressive
312 lattice strain evoked in austenite during quenching has relaxed, assuming that no change in
313 composition occurred. On cooling to 298 K (25 °C), no additional changes of $\varepsilon_\perp^{hkl\gamma}$ are observed.
314 Data is largely consistent with recent work in the literature [53], and indicates that residual stresses
315 in austenite are partially relaxed during tempering of martensitic stainless steel to 748 K (475 °C).

316 *Martensite*

317 At the beginning of the investigation, $a^{\alpha'} \approx a_{ref}^{\alpha'}$, which implies that $\varepsilon_\perp^{\alpha'} \approx 0$.

318 On heating, the measurement of $a^{\alpha'}$ shows four distinct regimes:

- 319 • for $T < 423$ K (150 °C), the rate of expansion of the unit cell of martensite is consistent with
320 $\lambda_{\alpha'}$;
- 321 • for the temperature range 423-748 K (150-475 °C), $a^{\alpha'}$ expands at a rate that varies with
322 hkl and in general is smaller than $\lambda_{\alpha'}$;
- 323 • for temperatures in the range 748-898 K (475-625 °C) a significant difference between $a^{\alpha'}$
324 and $a_{ref}^{\alpha'}$ yields a large deviation between the observed and predicted lattice parameter at
325 898 K (625 °C);
- 326 • at 898 K (625 °C), martensite becomes paramagnetic and $\lambda_{\alpha'}$ increases significantly.

327 On cooling, martensite reflections have sufficient intensity for a temperature of 403 K (130 °C),
328 when approx. 5 % of the austenite has transformed. For fractions up to $f_{\perp}^{\alpha} \approx 0.2$, data scatters
329 significantly. For martensite fractions beyond 0.2, the lattice strain in martensite varies significantly
330 depending on the probed hkl . On average, $a_{\perp}^{hkl\alpha'} < a_{ref}^{\alpha'}$ and about constant over the whole
331 transformation process.

332 During tempering, the evolution of $a_{\perp}^{\alpha'}$ on heating to 748 K (475 °C) is consistent with the data
333 obtained on heating the sample “as normalized” and the lattice of martensite expands less than
334 predicted based on $\lambda_{\alpha'}$ in the temperature interval 448-748 K (175-475 °C). This deviation is
335 retained on subsequent cooling to 298 K (25 °C).

336 Data acquired during the two heating steps indicates that tempering of martensite proceeds in two
337 stages in this alloy: the first stage extends from 423 K (150 °C) to 623 K (350 °C); the second stage
338 from 748 K (475 °C) to 898 K (625 °C). Data is consistent with previous work in the literature [53]
339 and could either be interpreted in terms of the introduction of compressive lattice strains, $\varepsilon_{\perp}^{hkl\alpha'}$, in
340 martensite or as the rejection of C and N from solid solution. Applying the unit cell volumes for Fe-
341 C and Fe-N martensite from Ref. [15],** and assuming that $\varepsilon_{\perp}^{hkl\alpha'}$ is not affected by tempering, the
342 observed differences between the variation of $a_{\perp}^{\alpha'}$ with temperature and $\lambda_{\alpha'}$ would correspond to the
343 rejection of approx. 0.03 wt% C+N during the first stage of tempering and 0.04 wt% C+N during
344 the second stage. These values are considered realistic taking into account the alloy composition (cf.
345 Table 1), and imply that the effect of tempering on $a_{\perp}^{\alpha'}$ can mainly have its origin in a change of the
346 content of interstitials in solid solution in martensite.

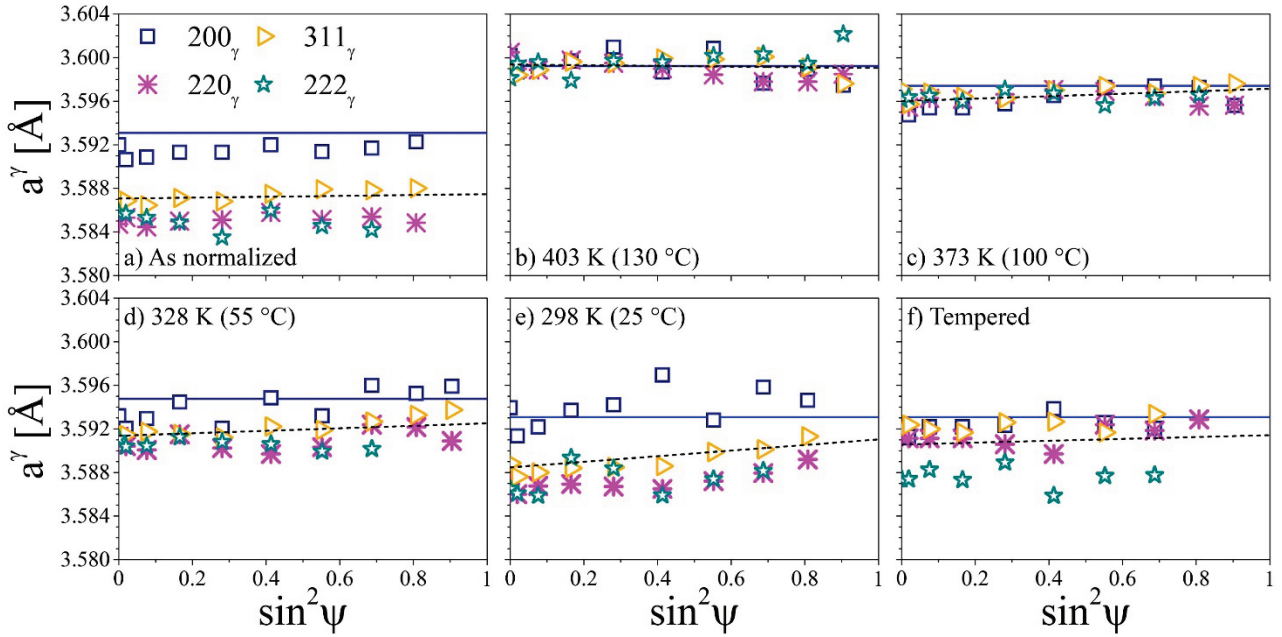
347 **3.3.State of stress – sample 1**

** Note that the relative difference between the effects of C and N (in wt%) on the unit cell volume of martensite is <3 %, and can be neglected within the experimental accuracy indicated in Ref. [15].

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3.3.1. "as normalized" condition

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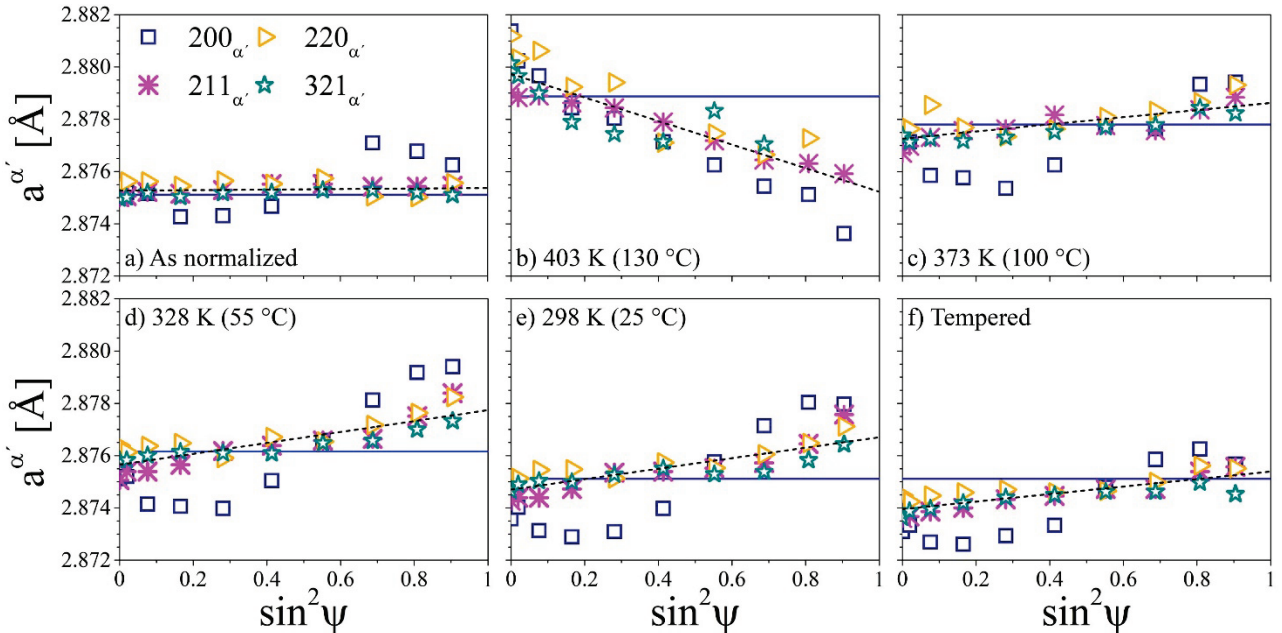
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Figure 4. Lattice parameter of austenite a^γ evaluated from 200_γ , 311_γ , 220_γ and 222_γ and plotted versus $\sin^2\psi$. Data acquired: (a) at the beginning of the investigation; (b) at 403 K (130 °C) during quenching; (c) at 373 K (100 °C) during quenching; (d) at 328 K (55 °C) during quenching; (e) at the end of the quenching cycle; (f) at room temperature after tempering to 748 K (475 °C). The blue continuous lines represent the strain-free lattice parameter of austenite a^γ_{ref} . The black dashed lines were obtained by linear regression among a^γ values averaged over the probed hkl .



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Figure 5. Lattice parameter of martensite a^α evaluated from 200_α , 211_α , 220_α and 321_α and plotted versus $\sin^2\psi$. Data acquired: (a) at the beginning of the investigation; (b) at 403 K (130 °C) during quenching; (c) at 373 K (100 °C) during quenching; (d) at 328 K (55 °C) during quenching; (e) at the end of the quenching cycle; (f) after tempering to 748 K (475 °C). The blue continuous lines represent the strain free lattice parameter of martensite a^α_{ref} . The black dashed lines were obtained by linear regression among a^α values averaged over the probed hkl , excluding 200_α .

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364 In the normalized condition the steel contains a martensite fraction $f^{\alpha'} = 0.95$; the rest is retained
365 austenite. Representative values of a^{γ} and $a^{\alpha'}$, determined from various hkl are presented as a
366 function of ψ in Figs. 4a and 5a, respectively. Some data points at high ψ angles were excluded
367 because the diffracted intensity was insufficient for accurate peak fitting.

368 Fig. 4a shows that, within experimental accuracy, a^{γ} is independent of ψ , indicating that the state
369 of stress is effectively hydrostatic (or, trivially, nil), but depends on hkl . Comparing a^{γ} with a_{ref}^{γ}
370 (the latter as represented by the solid blue line in Fig. 4a) reveals that austenite experiences a hkl
371 dependent compressive lattice strain. Compressive strain is largest for 220_{γ} and 222_{γ} , very small
372 for 200_{γ} , while compression for 311_{γ} is close to the value obtained from averaging over all
373 measured hkl . Stress analysis yields $\sigma_{\parallel}^{\gamma} - \sigma_{\perp}^{\gamma} \approx 0$ and σ_{\perp}^{γ} equal to -0.21 GPa, -1.22 GPa,
374 -0.85 GPa and -1.29 GPa for probing 200_{γ} , 220_{γ} , 311_{γ} and 222_{γ} , respectively, giving an
375 average stress value $\langle \sigma^{\gamma} \rangle = -0.89$ GPa. It is noted that the order of increasing lattice strain (and
376 associated elastic residual stress), coincides with an increase of the orientation parameter $3\Gamma = 3 \cdot$
377 $\frac{h^2k^2+k^2l^2+l^2h^2}{(h^2+k^2+l^2)^2}$, which varies from 0 for 200_{γ} to 1 for 222_{γ} and for 311_{γ} is about half-way the range
378 ($3\Gamma = 0.47$), consistent with 311_{γ} representing the average over all hkl . This would suggest that
379 the observed differences for the probed hkl s are a consequence of elastic anisotropy in austenite.
380 This contrasts with the zero slope in Fig. 4a., because for a hydrostatic state of stress no dependence
381 of (elastic) lattice strain over hkl would be expected. In fact, $\left[3 \cdot s_1^{hkl\phi} + \frac{1}{2}s_2^{hkl\phi} \right]$, cf. Eq.1, is
382 independent of hkl , as can be verified for the data in Table 2. Evidently, the state of stress in
383 austenite is hydrostatic within the probed volume, but not necessarily hydrostatic over the length
384 scale of a single austenite grain (cf. Refs. [19,20]).

385 Fig. 5a shows that $a^{\alpha'}$ evaluated from $211_{\alpha'}$, $220_{\alpha'}$ and $321_{\alpha'}$ neither varies with ψ nor with hkl
386 within experimental accuracy. In contrast, the $\sin^2\psi$ dependence of $a^{\alpha'}$ derived from $200_{\alpha'}$ is
387 characterized by oscillations (cf. Ref. [4]) and therefore excluded from the analysis. Stress analysis
388 based on $211_{\alpha'}$, $220_{\alpha'}$ and $321_{\alpha'}$ showed that $\sigma_{\parallel}^{\alpha'} - \sigma_{\perp}^{\alpha'} \approx 0$. The average phase specific
389 (hydrostatic) stress in martensite calculated with Eq.4 is $\langle \sigma^{\alpha'} \rangle = 0.04$ GPa.

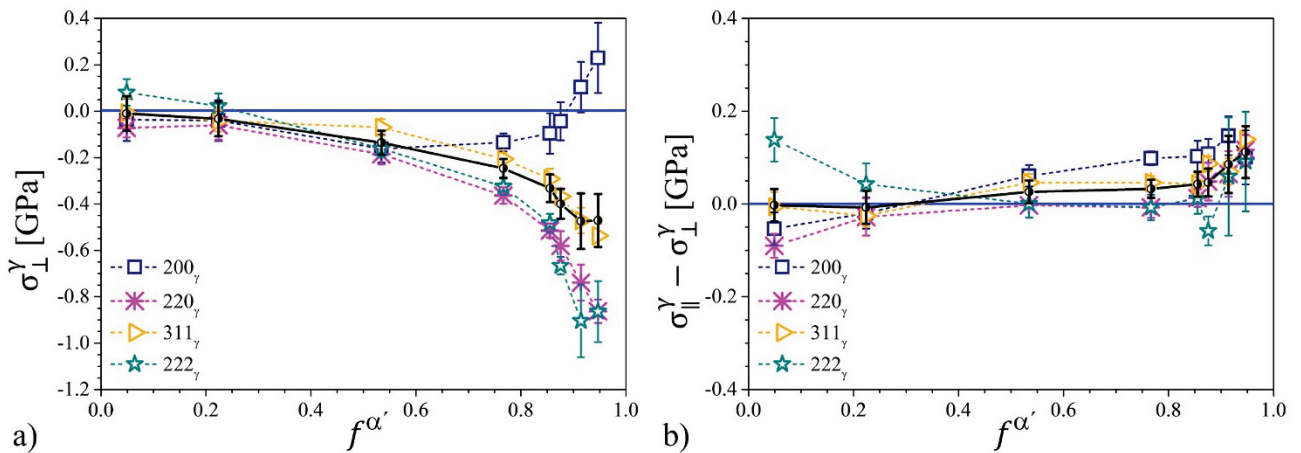
390 **3.3.2. Stress developing on martensite formation**

391 Representative examples of lattice parameter a_{ψ}^{ϕ} versus $\sin^2\psi$ data, as obtained with in situ
392 application of the $\sin^2\psi$ method, are reported in Figs. 4b-e and Figs. 5b-e. Stress values were
393 derived from such a_{ψ}^{ϕ} versus $\sin^2\psi$ relations, applying the XECs from Table 2. The stress values

394 obtained are given in Figs. 6 and 7 as a function of $f^{\alpha'}$. The fraction of transformed austenite was
 395 determined by averaging the values of $f_{\psi}^{\alpha'}$ measured at all the applied tilting angles, ψ . This
 396 procedure reduces significantly the uncertainty caused by crystallographic texture [46]. Figures 6a
 397 and 7a show the stresses in austenite and martensite as determined in the \perp direction. These stresses
 398 are interpreted as phase specific micro-stresses of type II. Figures 6b and 7b illustrate the difference
 399 between the stress components in the \parallel and \perp directions versus $f^{\alpha'}$. These stresses are interpreted as
 400 macro-stresses of type I, for austenite (Fig. 6a) and martensite (Fig. 7a), respectively.

401 From Fig. 4, it follows that as long as martensite is the minority phase in the sample, i.e. for $T \geq$
 402 388 K ($115 \text{ }^{\circ}\text{C}$), the lattice parameter for austenite is about the reference value, $a_{\psi}^{\gamma} \approx a_{ref}^{\gamma}$,
 403 implying that $\varepsilon_{\psi}^{hkl\gamma} \approx 0$ (Fig. 4b). At 373 K ($100 \text{ }^{\circ}\text{C}$), a small compressive lattice strain develops in
 404 austenite (Fig. 4c). This lattice strain increases on further cooling (Figs. 4c-e) and is most
 405 pronounced at 298 K ($25 \text{ }^{\circ}\text{C}$) (Fig. 4e). The lattice strain does not depend significantly on ψ , but it
 406 does vary with hkl . The variation among the various $\varepsilon_{\psi}^{hkl\gamma}$ increases during continuous cooling and,
 407 analogously, is most pronounced at 298 K ($25 \text{ }^{\circ}\text{C}$) (Fig. 4e). Stress analysis reveals that
 408 compressive stress of type II builds up in austenite for $f^{\alpha'} > 0.2$ (Fig. 6a). Compressive stress
 409 increases steadily with a reduction of the austenite fraction and depends on the $\varepsilon_{\psi}^{hkl\gamma}$ from which it
 410 is evaluated.

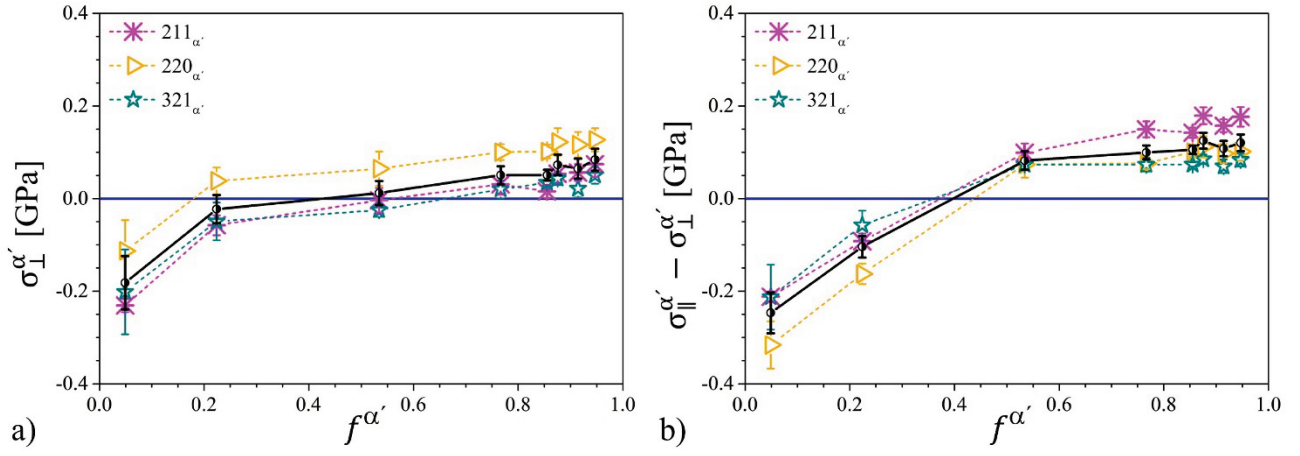
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413 **Figure 6. State of stress in austenite evaluated from lattice strains obtained for 200_{γ} , 220_{γ} , 311_{γ} and 222_{γ} and plotted**
 414 **versus the fraction of martensite formed $f^{\alpha'}$: a) stress component in the direction perpendicular to the sample surface σ_{\perp}^{γ} ; b)**
 415 **difference between the stress components parallel and perpendicular to the sample surface $\sigma_{\parallel}^{\gamma} - \sigma_{\perp}^{\gamma}$. The black lines and**
 416 **symbols represent the state of stress averaged over all probed reflections. Error bars indicate the standard error of the**
 417 **estimate for linear regression of data in Fig. 2. An additional experimental error related to the alignment of the**
 418 **diffractometer is estimated in the order of $\pm 30 \text{ MPa}$.**

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Figure 7. State of stress in martensite evaluated from lattice strains obtained for $200_{\alpha'}$, $211_{\alpha'}$, $220_{\alpha'}$ and $321_{\alpha'}$ and plotted versus the fraction transformed $f^{\alpha'}$: a) stress component in the direction perpendicular to the sample surface $\sigma_{\perp}^{\alpha'}$. b) difference between the stress components parallel and perpendicular to the sample surface $\sigma_{\parallel}^{\alpha'} - \sigma_{\perp}^{\alpha'}$. The black lines and symbols represent the state of stress averaged over all probed reflections apart from $200_{\alpha'}$. Error bars indicates the standard error of the estimate for linear regression of data in Fig. 2. An additional experimental error related to the alignment of the diffractometer is estimated in the order of ± 30 MPa.

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After quenching, σ_{\perp}^{γ} equals $+0.23$ GPa, -0.86 GPa, -0.54 GPa and -0.86 GPa when evaluated from 200_{γ} , 220_{γ} , 311_{γ} and 222_{γ} , respectively, and $\langle \sigma_{\perp}^{\gamma} \rangle = -0.51$ GPa. The atypical behaviour of 200_{γ} cannot be explained solely in terms of elastic anisotropy in austenite (see previous paragraph). This behavior is consistent with observations in the literature on the influence of plastic accommodation of the volume changes on the suitability of hkl for the determination of residual stresses in f.c.c. metals [29-32] and strongly suggests that plastic deformation has occurred in austenite during martensite formation.

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During the transformation, the state of stress in (untransformed) austenite is close to hydrostatic within the probed volume (i.e. $\sigma_{\parallel}^{\gamma} - \sigma_{\perp}^{\gamma} \ll \sigma_{\perp}^{\gamma}$). However, measurable macro-stress is present, as reflected by $\sigma_{\parallel}^{\gamma} \neq \sigma_{\perp}^{\gamma}$ (Fig. 6b). At the beginning of the transformation, $\sigma_{\parallel}^{\gamma} > \sigma_{\perp}^{\gamma}$ as evaluated from $\varepsilon_{\psi}^{222\gamma}$, which, among the probed γ reflections, offers the largest information depth. Probing the other hkl at shallower information depths it is found that $\sigma_{\parallel}^{\gamma} \leq \sigma_{\perp}^{\gamma}$. Along with an increase of the fraction of martensite, a small tensile macro-stresses, i.e. ($\sigma_{\parallel}^{\gamma} > \sigma_{\perp}^{\gamma}$), builds up for $f^{\alpha'} > 0.2$.

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Fig. 5 shows that $a_{\psi}^{\alpha'}$ was linearly dependent on $\sin^2\psi$ at $T = 403$ K (130 °C), i.e. just below M_s (Fig. 5b). The slope of $a_{\psi}^{\alpha'}$ versus $\sin^2\psi$ is negative. Upon cooling to $T=373$ K (100 °C), the slope of $a_{\psi}^{\alpha'}$ versus $\sin^2\psi$ reverts to positive (Fig. 5c) and a non-linear dependence of $a_{\psi}^{\alpha'}$ on $\sin^2\psi$ is observed for $200_{\alpha'}$, reflecting the oscillations in the $a_{\psi}^{\alpha'}$ vs. $\sin^2\psi$ distribution in the “as normalized” condition (Fig. 5a). On continuous cooling to 298 K (25 °C), $a_{\psi}^{\alpha'}$ shrinks at a rate

446 commensurate with the thermal expansion coefficient $\lambda_{\alpha'}$ and $\varepsilon_{\psi}^{hkl\alpha'}$ does not vary significantly
447 with hkl (Figs. 5c-e).

448 Stress evaluation reveals that martensite experiences an average compressive stress at the beginning
449 of the transformation, which reverts into an average tensile stress for $f^{\alpha'} > 0.5$ (Fig. 7a). The state
450 of stress is not particularly sensitive to the hkl used for probing the lattice strain. Finally, after
451 quenching, $\langle \sigma_{\perp}^{\alpha'} \rangle = +0.08 \text{ GPa}$.

452 Fig. 7b also indicates the presence of macro-stresses. At the beginning of the process, macro-
453 stresses are compressive and significant, approx. -0.25 GPa . With increasing martensite content,
454 compression decreases and is reversed into tensile macro-stress for $f^{\alpha'} > 0.2$.

455 3.3.3. The tempered condition

456 The values of $a_{\psi}^{hkl\gamma}$ and $a_{\psi}^{hkl\alpha'}$ are shown as a function of $\sin^2\psi$ in Figs. 4f and 5f, respectively.

457 The fraction of martensite in the sample is $f^{\alpha'} = 0.95$, consistent with the “as normalized” and “as
458 quenched” conditions. The result in Fig. 4f shows that $a_{\psi}^{hkl\gamma}$ varies with hkl , but not with ψ .

459 Comparison of a^{γ} with a_{ref}^{γ} reveals that lattice strain evaluated from 200_{γ} and 311_{γ} is negligibly
460 small, whereas $\varepsilon^{222\gamma} \ll 0$. Stress analysis shows that σ_{\perp}^{γ} is -0.07 GPa , -0.23 GPa , -0.08 GPa and
461 -0.86 GPa when evaluated based on 200_{γ} , 220_{γ} , 311_{γ} and 222_{γ} , respectively. The stress is
462 approximately hydrostatic within the probed volume, with $\langle \sigma_{\perp}^{\gamma} \rangle = -0.31 \text{ GPa}$ and $\sigma_{\parallel}^{\gamma} - \sigma_{\perp}^{\gamma} =$
463 $+0.04 \text{ GPa}$. The hkl -dependent lattice strain suggests that a hydrostatic state of stress does not
464 apply at the length scale of a single austenite grain.

465 Fig. 5f shows that $a_{\psi}^{hkl\alpha'}$ evaluated from $211_{\alpha'}$, $220_{\alpha'}$ and $321_{\alpha'}$ is a linear function of $\sin^2\psi$ but
466 does not vary with hkl ; $a_{\psi}^{200\alpha'}$, instead, shows a $\sin^2\psi$ dependence characterized by oscillations as
467 earlier reported for the material in “as normalized” and “as quenched” conditions. Stress analysis
468 was based on $211_{\alpha'}$, $220_{\alpha'}$ and $321_{\alpha'}$. Under the assumption that the chemical composition of the
469 martensite has not changed during tempering it is obtained $\langle \sigma_{\perp}^{\alpha'} \rangle = -0.09 \text{ GPa}$ and $\sigma_{\parallel}^{\alpha'} - \sigma_{\perp}^{\alpha'} =$
470 $+0.08 \text{ GPa}$.

471 Nevertheless, the material is almost fully martensitic during tempering. A significant variation of
472 $\langle \sigma_{\perp}^{\alpha'} \rangle$ from $\langle \sigma_{\perp}^{\alpha'} \rangle = 0.08 \text{ GPa}$ before to tempering to $\langle \sigma_{\perp}^{\alpha'} \rangle = -0.09 \text{ GPa}$ after tempering would
473 imply an enormous (about 3 GPa) balancing variation in $\langle \sigma_{\perp}^{\gamma} \rangle$, (cf. Eq.7), which is not confirmed by
474 the experimental data. Evidently, the shrinkage of the martensite lattice from the “as quenched” to
475 the “tempered” condition is not due to a change of $\langle \sigma_{\perp}^{\alpha'} \rangle$, but has its origin in a change in chemical

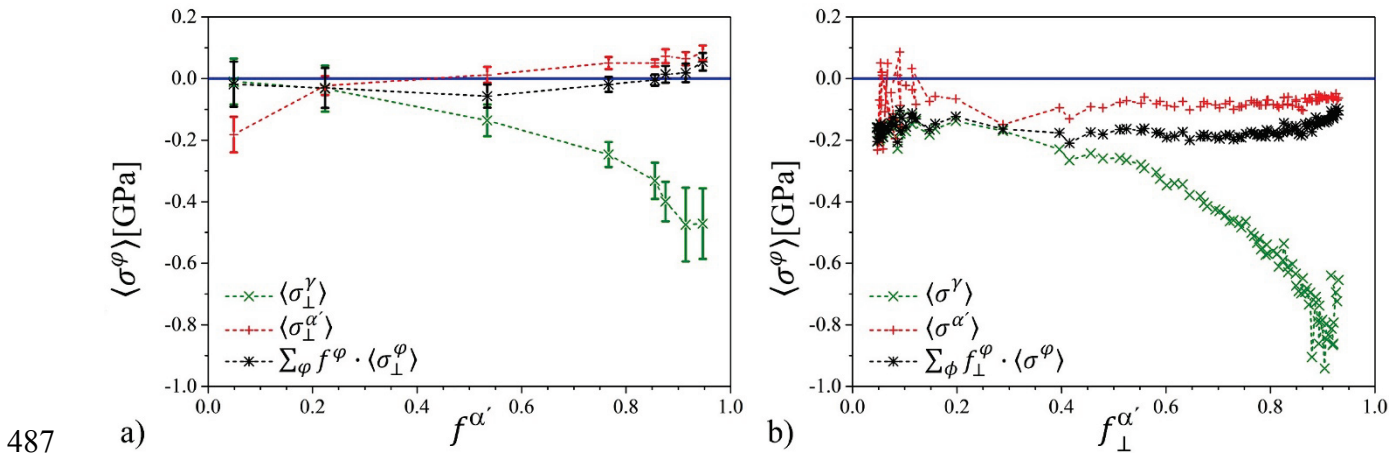
476 composition. The measured change in the lattice parameter corresponds to expelling approx. 0.03
 477 wt% C+N from solid solution in martensite during tempering to 748 K (475 °C).

478 4. Discussion

479 4.1. Partitioning of stresses in austenite and martensite during martensite formation

480 Historically, the evolution of the phase specific stresses, $\langle \sigma^\varphi \rangle$, in austenite and martensite during
 481 the austenite-to-martensite transformation has been investigated by measuring the evolution of the
 482 phase specific strain, ε^φ , as a function of the corresponding phase fraction, f^φ , either in a single
 483 direction [14,16-18,22,23], or in a set of directions (approx.) normal to the incident beam
 484 [19,20,27]. These investigations departed from the hypothesis that macro-stresses are negligible
 485 within the volume probed by XRD.

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488 **Figure 8.** Calculated phase-specific stresses $\langle \sigma^\varphi \rangle$ for a) sample 1 and b) sample 2. Data re plotted versus fraction transformed
 489 $f^{\alpha'}$ (averaged over all ψ) and fraction transformed evaluated in the direction normal to the sample surface f^α_\perp , for sample 1
 490 and sample 2, respectively. Error bars indicates minimum and maximum values based on data in Figs. 6a and 7a. An
 491 additional experimental error related to the alignment of the diffractometer is estimated in the order of ± 30 MPa. Error
 492 analysis do not consider uncertainty in the determination of $f^{\alpha'}$ and f^α_\perp .

493

494 In the present work, the evolution of stress in the material during martensite formation including the
 495 separation of macro- and micro-stresses was investigated in situ for the first time (sample 1). To
 496 verify whether the measured lattice strains in austenite can be ascribed to (micro-)stresses of type II,
 497 a balancing of average stress values in the phases in the sample (cf. Eq. 7) with $f^{\alpha'}$ was considered.
 498 The results of this verification for the stress values presented in Fig. 6a and 7a are shown in Fig. 8a.

499 Fig. 8a shows that, within experimental accuracy, internal balance of average stresses is satisfied
 500 from the onset of the transformation to its completion. At the beginning of the transformation
 501 martensite is the minority phase and experiences significant phase specific compressive (micro-

502)stress of type II, which is balanced by very small tensile (micro-)stress of type II in the dominating
503 austenite. During martensite formation, the state of stress in the phases reverses. For $f^\alpha > 0.2$,
504 tension builds up in martensite and compression develops in austenite. At the end of the
505 transformation, significant compressive (micro-)stress of type II in austenite is balanced by small
506 tensile (micro-)stress of type II in martensite. Data does not provide a reason for the observed
507 reversion of the state of stress in the phases during transformation.

508 The importance of performing in situ stress analysis to evaluate the state of stress in the phases is
509 clarified from comparing Fig. 8a and Fig. 8b. The stress values presented in Fig. 8b (sample 2) were
510 obtained applying Eq. 2, under the assumption that the state of stress in both phases is hydrostatic.
511 Fig.8b indicates that martensite formation leads to compressive stresses in both martensite and
512 austenite, in agreement with a previous claim in Ref. [14]. The in situ stress analysis as determined
513 for sample 1 demonstrates that this conclusion is incorrect. Evidently, Eq.2 can be applied only if
514 the assumption that macro-stresses are negligible is validated.

515 Macro-stresses can be introduced as a consequence of a variation in the degree of transformation
516 over the sample thickness. It is well accepted that martensite formation starts at the surface and is
517 associated with a volume expansion. This expansion is partially relaxed in the \perp direction and
518 partially accommodated within the sample. Martensite, which is concentrated at the sample surface,
519 experiences compressive macro-stresses (Fig. 7b, left), which are balanced by tensile macro-stresses
520 in the bulk austenite, as reflected in Fig. 6b (left) by probing the 222_γ reflection. Thereafter,
521 continuous cooling promotes continuation of the transformation (from left to right in Figs. 6 and 7),
522 including transformation of the bulk. Transformation of the bulk, is similarly associated with a
523 volume expansion. However, the expansion of the bulk is fully counteracted by the martensitic case,
524 which surrounds it. As a result, macro-stress in the near surface region changes from compressive to
525 tensile during continued transformation, as evidenced in Figs. 6 and 7 at $f^{\alpha'} > 0.2$.

526 Additionally, hkl dependent lattice strain in austenite should be addressed. Historically, the lattice
527 strain in austenite, ε^γ , developing during martensite formation was obtained from either a^γ
528 evaluated from the position of a single reflection [11-15,23,24], or from an average a^γ value
529 obtained from the simultaneous evaluation of the position of all probed reflections [19,20,27]. In a
530 few studies [16,17,22], hkl -specific $\varepsilon^{hkl\gamma}$ relying on an independent evaluation of corresponding
531 $a^{hkl\gamma}$ were presented. These latter studies revealed that the lattice strain is compressive ($\varepsilon^{hkl\gamma} < 0$)
532 and largest for $111_\gamma/222_\gamma$ [16,17,22], moderately negative and close to the average value for 311_γ
533 [16,17] and negligible [16,17] or positive, for 200_γ [22]. These observations are confirmed by the
534 results obtained in the present investigation. The hkl -dependence of lattice strain in austenite can

535 partly be explained from the anisotropic elastic properties of austenite (see section 3.3.). However,
536 lattice strains of opposite sign for different hkl , as observed for the material as quenched and as
537 tempered, cannot be reconciled with elastic anisotropy only. Plastic accommodation of the
538 transformation strain is considered responsible for this behavior. When a unit of martensite forms,
539 the transformation evokes tension in the surrounding austenite, which yields heterogeneously.
540 Crystal plasticity in f.c.c. crystals yields hkl dependent non-linear stress-strain behaviour in addition
541 to elastic anisotropy and consequently, a change of the apparent elastic constants. As a result,
542 anisotropic residual stress remains after unloading. Unloading of the state of tension in austenite
543 takes place during continued martensite formation, as revealed by the development of an average
544 compressive state of stress in this phase.

545 Unfortunately, controversy exists as to which hkl s should be chosen to prevent these anisotropy
546 effects (cf. Ref. [29-31] vs. Ref. [32]). If only elastic anisotropy is responsible for hkl dependence,
547 averaging is effectively obtained by choosing the 311_γ reflection, as the corresponding orientation
548 parameter 3Γ for this reflection is close to 0.5, i.e. half way the range from 0 to 1. In the present
549 work, $\langle\sigma^\gamma\rangle$ was taken as the average value for stress obtained from lattice strain over ε^{200_γ} , ε^{220_γ} ,
550 ε^{311_γ} and ε^{222_γ} . This procedure yielded the conclusion that Eq. 3 was satisfied throughout the
551 whole transformation process. It is explicitly mentioned that choosing the 311_γ reflection would
552 have given a comparable result.

553 Further insight in the elasto-plastic interaction between the phases during transformation is provided
554 by data collected for the ψ -dependent lattice parameter $a_\psi^{200\alpha'}$. A rigorous treatment to interpret a d
555 vs. $\sin^2\psi$ distribution characterized by oscillations is missing [4]. However, in a cubic phase,
556 oscillations in the d -vs. $\sin^2\psi$ distribution can be caused by texture and/or plastic strain. In textured
557 elastically strained cubic materials, no oscillation of $a_\psi^{\alpha'}$ versus $\sin^2\psi$ is expected for the $h00_{\alpha'}$
558 reflections [4,54]. On the other hand, oscillations will arise in plastically strained crystals, and will
559 be most significant for $200_{\alpha'}$ [4,55]. Hence, the present data indicates that both austenite and
560 martensite are plastically strained during the transformation.

561 Finally, we suggest that a consistent description of the evolution of strain and phase-specific
562 stresses in the material during the austenite-to-martensite transformation requires that the elasto-
563 plastic interaction of the two phases and the elasto-plastic anisotropy of the two crystal lattices is
564 taken into account. Developing such description is beyond the scope of the present work.

565 **5. Conclusion**

566 Stress analysis was applied to investigate in situ stresses of type II evoked by the austenite-to-
567 martensite transformation in steel. The analysis shows indeed that such stresses do build up.
568 Stresses of type II have a significant magnitude for the minority phases, *i.e.* for martensite at the
569 beginning of the transformation and for austenite at the end.
570 Stresses of type II in the minority phase are compressive and are balanced by small tensile stresses
571 of type II in the majority phase.
572 Strain in austenite is anisotropic: maximum compression is observed for spacings of the $(222)_\gamma$ and
573 $(220)_\gamma$ planes, while tension was revealed in the $[200]_\gamma$ direction. Anisotropic strain in austenite is
574 particularly significant in the latest stage of the transformation.
575 Strain anisotropy in austenite and a d-vs. $\sin^2\psi$ distribution characterized by oscillations in
576 martensite indicate that both phases are subjected to plastic deformation during quenching.
577 An in-depth evaluation of the state of stress in the phases requires further investigation in the effect
578 of plasticity on the determination of stresses in iron-based alloys.
579 Tempering of soft martensitic stainless steel to 748 K (475 °C) yields partial relaxation of stresses.

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