This is an electronic version of an article whose final and definitive form has been published in Scripta Mater. **64**, 466-9 (2011).
Scripta Materialia is available online at: http://www.sciencedirect.com/.

# ON ABNORMAL GROWTH OF GOSS GRAINS IN GRAIN ORIENTED SILICON STEEL

A.Morawiec

Polish Academy of Sciences
Institute of Metallurgy and Materials Science
Reymonta 25, 30–059 Kraków, Poland
E-mail: nmmorawi@cyf-kr.edu.pl
Tel.: ++48–122952854, Fax: ++48–122952804

ABSTRACT: A partial explanation of the phenomenon of abnormal growth of Goss oriented grains in silicon iron is proposed. It follows from an observation that texture evolves toward Goss orientation if grains of low surface energy have high probability of growth. A corresponding simulation starts with primary recrystallization texture and results in strong Goss texture. To explain development of early secondaries in subsurface layers, an option is considered that besides capillary forces, the growth is caused by surface induced lattice instabilities.

*Keywords*: iron alloys, soft magnets, secondary recrystallization, recrystallization texture, simulation

Grain oriented silicon iron (Fe ∼3%wt Si) is a conventional material for cores of electric transformers [1, 2]. Although the use of amorphous and nanocrystalline soft magnetic alloys is increasing, the 'GO electrical steels' still play a very significant role [3]. Among factors determining their good magnetic properties is very sharp texture dominated by the $\{110\}<001>$ (Goss) component. There are a number of processing routes leading to the Goss texture (two or one cold rolling + annealing cycles, MnS or AlN as inhibitors of normal growth) [4]. Our considerations are limited to the phenomena that follow primary recrystallization, and the mechanism of this last phase of processing is believed to be the same for all routes. Briefly, this stage is a high temperature annealing of decarburized primary recrystallized material ($\sim 0.30mm$ thick sheet) with the initial grain size of about $20\mu m$, impurities in the form of dispersed–phase particles, and relatively weak texture consisting mainly of the $\{111\}<uvw>$ ($\gamma$) and $\{hk0\}<001>$ ($\eta$) fibers. The texture is slightly inhomogeneous with the $\eta$ fiber stronger in the subsurface layers than at the sheet center; e.g., [5]. The annealing leads to secondary recrystallization: normal grain growth is suppressed by precipitates while some grains (secondaries) grow abnormally to large dimensions.

The control of the sharpness of the Goss texture is crucial for the quality of the final product. Progress in optimizing the texture is possible through understanding of the processes involved. Thus, questions arise about the mechanism of the exaggerated growth of grains and the cause of the selection of the particular Goss orientation. These issues have not been explained despite intense research in this area. We will not make any attempt to review the vast literature of the subject. Let us only note that numerous theories have been proposed and then abandoned (e.g., size advantage of Goss oriented grains, greater perfection of their interior, suitably oriented strains directing orientation changes). One of the lasting postulates is that the secondary recrystallization is driven by capillary forces, i.e., that interfacial (surface and/or boundary) tension provides energy for boundary migrations, which change proportions of texture components. Philip and Lenhart [6] noticed that early secondaries are located under the surface inside the specimen; this led to the conclusion that grain boundary energy, and not surface energy, is the actual driving force. Currently, a common view is that "secondary recrystallization is a process of nucleation followed by grain growth. The driving force for this process is the reduction of grain boundary energy" [7, 8]. All recent theories (high mobility of CSL boundaries [9, 10] or high energy boundaries [11] caused by preferential particle coarsening, solid state wetting [12]) are based on the above principle, with special properties ascribed to boundaries characterized by particular grain misorientations. However, the misorientation–based models do not explain the sharpness of the final texture: in such models, the selection process is determined by the orientations of the primary grains, and the spread of these orientations is large; cf. [13, 14]. Also the principle that growth is driven by grain boundaries becomes problematic in confrontation with the intricate early stage of secondary recrystallization, when Goss grains are in the minority and not larger than other grains. To describe this phase, the principle must be augmented, and an additional driving force needs to be indicated.

Below, an explanation of the rise of the Goss component is proposed. Briefly, it is shown that growth of grains with $\{hk0\}$ planes parallel to the rolling plane, with particular emphasis on $\{110\}$, leads to the Goss texture. An important characteristic of the $\{hk0\}$ planes, in particular $\{110\}$ planes, is their low surface free energy. Therefore, the texture evolution toward the Goss orientation follows, if the orientation changes are assumed to be influenced by the surface of the sheet. It will only remain to clarify the mechanism of the transmission of the changes to other grains. It is possible that driving force coming from the reduction of interfacial energy may work in combination with other mechanisms of lattice reorientations. With the involvement of surfaces, the explanation partly departs form the keystone assumption of recent theories that the process is determined by grain boundaries. The main aspects of the proposed growth scheme are considered below in more detail.

Let us begin with the issue of texture changes. It is known that the primary texture initially evolves toward the $\eta$ fiber, and only later the Goss component gets increasingly sharper; e.g., [5]. In light of this, the following observations are of interest: If only grains with $\{hk0\}$ planes close to the rolling plane have high probability of growth, the resulting texture will be dominated by the $\eta$ fiber because there are no other primary texture components of the type $\{hk0\}<uvw>$. Moreover, if only grains with $\{110\}$ planes parallel to the rolling plane have high probability of growth, the resulting texture will be dominated by the Goss component because there are no other significant $\{110\}<uvw>$ components in the primary texture. Consequently, these two theoretical facts are sufficient for faithful modeling of the actual texture evolution.

The above statements have been confirmed by a simple computer simulation. The simulation was based on the frequency of occurrence of particular orientations without spatial relationships between grains. In effect, it involved only texture induced correlations between orientations of grains. In each simulation step, a randomly selected 'grain' grew at the expense of another 'grain'. The probability of growth depended on 'grain' orientation with respect to the rolling plane. For a given grain, it was, respectively, 50% or 45% if one of its $\{110\}$ or other $\{hk0\}$ planes was within 7° to the rolling plane; otherwise, the probability was only 10%. These probabilities agree with with the rules specified in the previous paragraph. At the outset of the simulation, there were $N = 3 \times 10^6$ 'grains' of equal size. Their orientations were generated in such a way that the orientation distribution corresponded to an experimental primary recrystallization texture (taken from [15]). The total number of 'grains' declined with the progress of the simulation process. After some steps, the texture contained mostly $\eta$ fiber, and with a sufficiently large number of steps, Goss became the only strong component; see Fig. 1. Thus, the texture changes in the simulation are analogous to those occurring during secondary recrystallization, and these results substantiate the theoretical claims made above. The simulation is stable with respect to the numbers (probabilities of growth) used; different values give different texture intensities but the trends are the same. We are not aware of any other simulations based on similarly simple postulates, which would mimic the experimental texture evolution so closely.

The crystallographic planes pointed out in previous paragraphs correspond to low surface free energy. Although experimental data on surface energy are strongly influenced by the environment, impurities, adsorption and diffusion from the bulk, there are indications that the energy has the lowest values at the $\{110\}$ and $\{100\}$ planes [16–18]. Results of theoretical calculations also vary considerably but data for iron hint that surface energies corresponding to $\{110\}$ and other low index $\{hk0\}$ planes are low compared to those of $\{111\}$ and high index planes; see, e.g., [19, 20].

The differences in the surface free energies of particular crystallographic planes are proven to play a leading role in secondary recrystallization in thin–gauge specimens of high purity Fe–Si [21, 22]. It is likely that the surface energy also plays a part in the secondary recrystallization in the presence of second phase particles. Moreover, there is a general argument for the involvement of the surface: for Goss selection, the orientation changes must be governed by a factor strongly linked to the sample coordinate system, and besides the rolling direction, this system is determined by planes parallel to the surface.

Based on the above observations, it is clear that the texture evolution could be explained by the growth driven by surface free energy. This idea, however, is inconsistent with aforementioned conclusion of Philip and Lenhart [6] and with similar subsequent observations that early secondaries occupy the *sub*surface layer, and the advancement of change along the very surface is retarded, e.g., [23]. Therefore, one cannot rely on the conventional growth model with low surface energy driving boundaries which come into contact with the surface, and one must allow for other growth mechanisms to be involved. We are not at the position to indicate a clear solution to this issue. Let us only mention that the orientation changes may not necessarily start at (rare) Goss nuclei, and proceed by the reduction of interface energy. They may, for instance, be caused by lattice instabilities similar to those preceding diffusionless structural transformations, with mechanical energy necessary for atomic displacements provided by thermal vibrations; see, e.g., [24]. Their amplitudes in "soft" directions can be sufficiently large to cause displacements resulting in a more stable reoriented lattice. Presence of such directions is implied by the high elastic anisotropy, which drastically grows with temperature (Fig. 2). On the phase diagram, the stability of the $\alpha$ phase is related to the distance to the $\gamma$ loop. The actual distance may be small at some locations in the material due to solute segregation or dissolution of precipitates, and the orientation changes can be triggered by local instabilities[1]. In the considered case, the first destabilizing factor is believed to be the sheet surface. The scenario involving instability–caused reorientations seems to be confirmed by the kinetics of orientation changes: Experiments demonstrate that secondary recrystallization is not a smooth process but the steady growth is accompanied by burst motions [4, 28]. It is also clearly visible that some grains become engulfed, and growth fronts are very different from those corresponding to minimization of boundary area.

With the above observations in mind, one can sketch a possible mechanism of Goss selection. It is believed that orientation changes toward the $\eta$ fiber and Goss orientation are initiated at some properly oriented grains on the surface, and then the changes penetrate to the subsurface layer and spread out there. First, crystals with low surface energy $\{hk0\}$ planes appear or grow on the surface, and this enhances the $\eta$ fiber. With the $\{110\}$ cusp of the surface energy deeper than the other $\{hk0\}$ cusps, the Goss grains ultimately dominate the texture via the reduction of the surface energy. Particular aspects of the process are subject of speculation. Besides the conventional growth driven by capillarity forces, the changes of orientations may be caused by lattice instabilities induced at the surface or instigated at grain boundaries by newly reoriented neighbors.

Summarizing, the key conclusion of this note is that the texture evolution and the sharpness of final texture in grain oriented silicon iron can be explained if grains with low surface free energy have high probability of growth. The proposed description of the phenomenon of abnormal growth of Goss grains is intended to circumvent deficiencies of the theories based purely on properties of grain boundaries. However, only some concepts were outlined, and many gaps need to be filled. The main questions concern: – actual roles played by the 'reduction of surface energy', 'reduction of boundary energy' and 'lattice instability' mechanisms, – the character of the transmission of the change to subsequent grains, – the type of atomic displacements responsible for the orientation changes, – the state of the surface and the impact of surface segregation on surface energy, – the impact of coatings and annealing atmosphere, – the influence of the sheet thickness, – the kinetics of the progress of growth toward the sheet center. Properly designed experiments may test the validity of the proposed hypothesis and clarify the above issues.

---

[1] The underlying reasons for such instabilities are anisotropy and anharmonic behavior of the lattice [27].

## REFERENCES

[1] N.P. Goss, Trans. Am. Soc. Met. 23 (1935) 511.

[2] B.D. Cullity, Introduction to Magnetic Materials, Addison–Wesley, Reading, 1972.

[3] H.A. Davies et al., J. Magn. Magn. Mater. 320 (2008) 2411.

[4] M. Matsuo, ISIJ Int. 29 (1989) 809.

[5] M.Z. Quadir, C.S.T. Chang, B.J. Duggan, ISIJ Int. 50 (2010) 264.

[6] T.V. Philip, R.E. Lenhart, Trans. Met. Soc. AIME 221 (1961) 439.

[7] N. Chen, S. Zaefferer, L. Lahn, K. Günther, D. Raabe, Acta Mater. 51 (2003) 1755.

[8] Z. Xia, Y. Kang, Q. Wang, J. Magn. Magn. Mater. 320 (2008) 3229.

[9] J.Harase, R.Shimizu, Acta Metall. Mater. 38 (1990) 1395.

[10] Y. Yoshitomi, Y. Ushigami, J. Harase, T. Nakayama,H. Masui and N. Takahashi, Acta metall. mater. 42 (1994) 2593.

[11] Y. Hayakawa, J. Szpunar, Acta Mater. 45 (1997) 4713.

[12] H. Park, D.Y. Kim, N.M. Hwang, Y.C. Joo, C.H. Han, J.K. Kim, J. Appl. Phys. 95 (2004) 5515.

[13] A.Morawiec, Scripta Mater. 43 (2000) 275.

[14] A.L. Etter, T. Baudin, R. Penelle, Scripta Mater. 47 (2002) 725.

[15] A. Morawiec, J.A. Szpunar, D.C. Hinz, Acta Metall. Mater. 41 (1993) 2825.

[16] B. Mills, M. McLean, E.D. Hondros, Phil. Mag. 27 (1973) 361.

[17] J.L. Walter and C.G Dunn, Acta Metall. 8 (1960) 497.

[18] W.L. Elban, M.A. Hebbar, J.J. Kramer, Metall. Trans. A 6 (1975) 1929.

[19] M.J.S. Spencer, A. Hung, I.K. Snook, I. Yarovsky, Surf. Sci. 513 (2002) 389.

[20] J.M. Zhang, D.D. Wang, K.W. Xu, Appl. Surf. Sci. 252 (2006) 8217.

[21] J.L. Walter, Acta Metall. 7 (1959) 424.

[22] J.L. Walter and C.G. Dunn, Trans. Metall. Soc. AIME 218 (1960) 1033.

[23] N.C. Pease, D.W. Jones, M.H.L. Wise, W.B. Hutchinson, Met. Sci. 15 (1981) 203.

[24] T.R. Finlayson, Aust. J. Phys. 36 (1983) 553.

[25] J. Maia Alves, A.M. Vallêra, J. Magn. Magn. Mater. 157–158 (1996) 378.

[26] J.L. Routbort, C.N. Reid, E.S. Fisher, D.J. Dever, Acta Metall. 19 (1971) 1307.

[27] P.C. Clapp, Phys. Stat. Sol. B 57 (1973) 561.

[28] H. Homma, B. Hutchinson, Acta Mater. 51 (2003) 3795.

## CAPTIONS

Figure 1: (a) Experimental distribution of orientations used as the starting point for the simulation. The $\varphi_1$–projection is used. (b) Distribution of orientations after $20 \times N$ steps (1154119 'grains'); mainly $\eta$ fiber. (c) Distribution of orientations after $1000 \times N$ steps (65733 'grains'); mainly Goss component. (d) Values of the orientation distribution at Goss orientation and representatives of $\eta$ and $\gamma$ fibers versus (number of steps)$/N$.

Figure 2: Schematic illustration of the dependence of Zener's anisotropy index $A$ on temperature for Fe 2.1 wt% Si. The curve is based on data reported in [25]. Since the anisotropy grows with Si content [26], the values of $A$ for the considered material are higher.

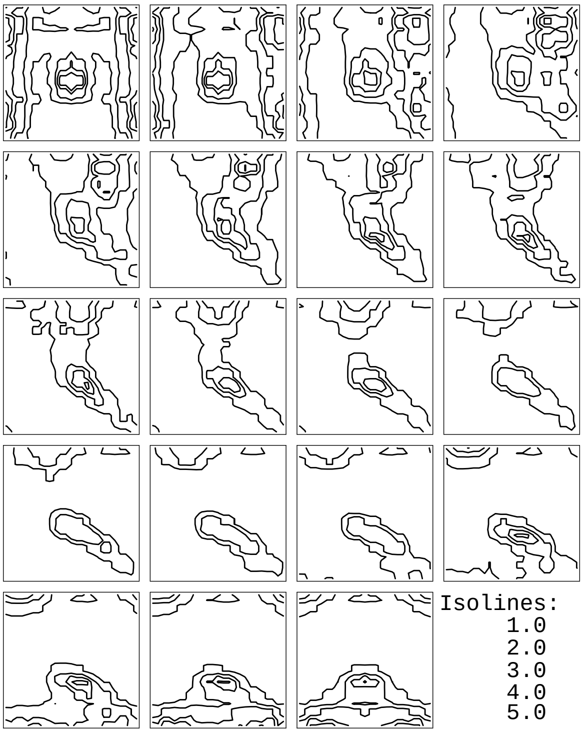

Fig. 1a

*A.Morawiec*, ... growth of Goss grains ...

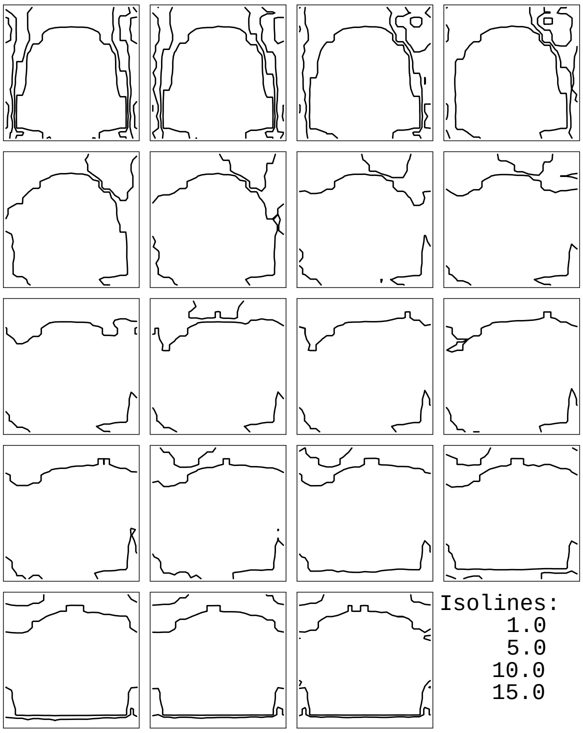

Fig. 1b

*A.Morawiec*, ... growth of Goss grains ...

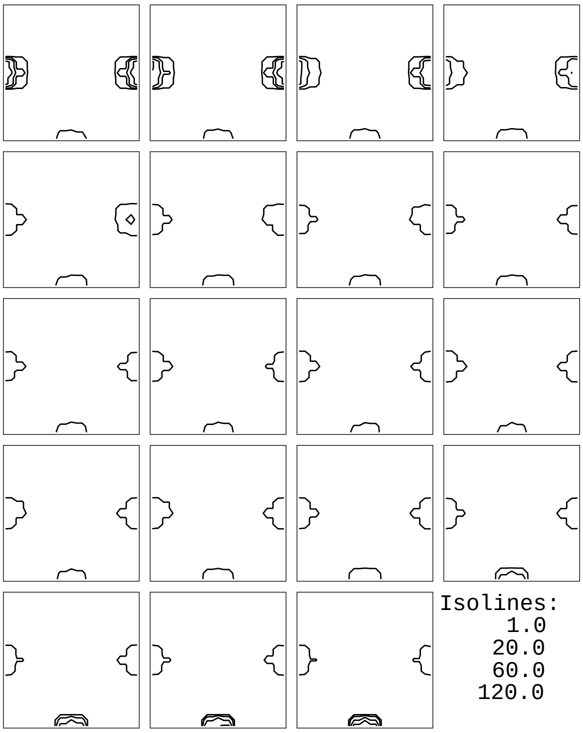

Fig. 1c

*A.Morawiec*, ... growth of Goss grains ...

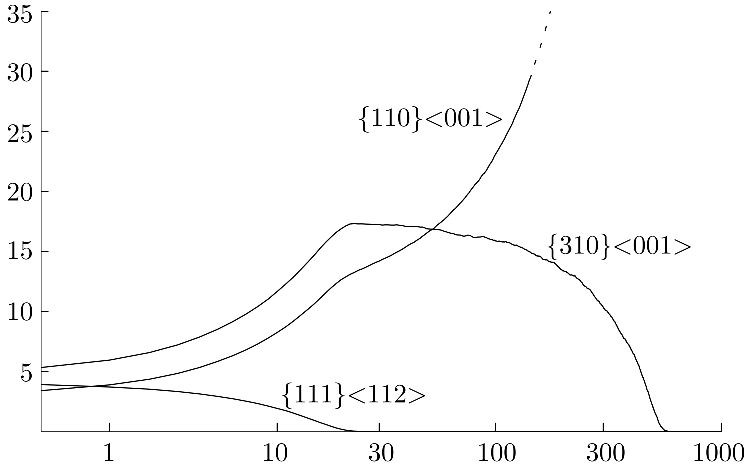

Fig. 1d

*A.Morawiec*, ... growth of Goss grains ...

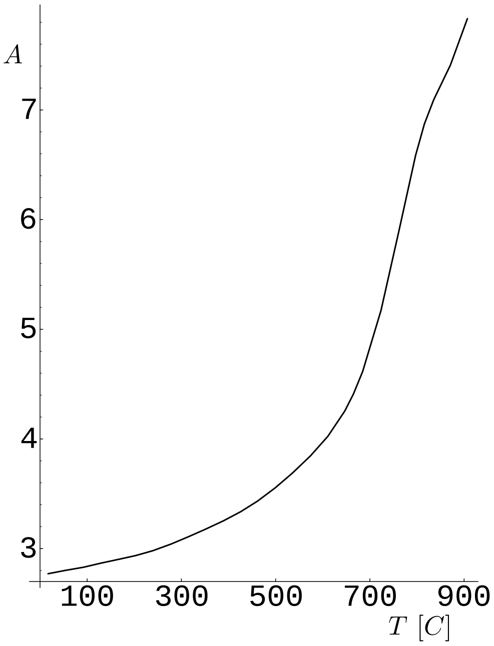

Fig. 2

*A.Morawiec*, ... growth of Goss grains ...