Interplay of dislocation-related relaxation processes in iron-based materials

M. J. Konstantinović*
Studiecentrum voor Kernenergie/Centre d'Etude de l'Energie Nucléaire (SCK·CEN), Boeretang 200, B-2400 Mol, Belgium
(Received 4 December 2007; revised manuscript received 12 June 2008; published 28 August 2008)

The internal friction spectra in various cold-worked iron-based alloys show that the intensity ratio of the peaks, corresponding to the relaxation processes at 0.52 eV (β peak) and 0.96 eV (γ peak), is proportional to the impurity concentration. Such correlated behavior of the two most dominant modes in the spectra is explained by a model which incorporates an increase in the β-relaxation process at the expense of the kink-antikink pair (γ-peak) nucleation process in pinned dislocation segments. Very good agreement between measured and calculated spectra is obtained by varying a single parameter: the ratio between the average size of the kink-antikink pairs and the average impurity-impurity distance.

DOI: 10.1103/PhysRevB.78.052104 PACS number(s): 61.72.Hh, 62.40.+i

I. INTRODUCTION

More than 50 years ago, Bordoni[1] demonstrated that the motion of dislocations in a periodic lattice potential,[2] occurring due to the discontinuous nature of the crystal lattice, leads to a thermally activated relaxation process. The internal friction spectra as a function of temperature in cold-worked fcc metals are found to exhibit a complex structure with a main maximum at approximately one-third of the Debye temperature.[1] Likewise, the internal friction measurements of the plastically deformed bcc metals show the existence of similar features (γ peak) but with much richer spectra due to the contribution of both screw and nonscrew types of dislocations in the relaxation process.[3] Moreover, in both types of materials, the motion of dislocations is governed by nucleation of the kink-antikink pairs (kink pairs),[4] which ensures that the dislocation segment can overcome the Peierls barriers and relieve the stress in the material. The critical stress associated with the kink-antikink nucleation mechanism in screw dislocation motion is proved to be crucial for determination of the kink-pair energy.[5] The Seeger-Schiller model and subsequent developments/improvements are still used to tackle the most obscure experimental observations, such as the origin of subsidiary peaks, observed in the internal friction spectra of both fcc and bcc metals.[6] These peaks, usually referred to in the literature as Niblett-Wilks (NW) peaks in fcc metals[7] and as β peaks in bcc metals,[8] also appear under plastic deformation and decrease upon annealing, indicating close relationship with the relaxation process due to the kink-pair generation. However, there is a dispute in the literature about the activation mechanism responsible for these relaxation processes. In fcc metals, according to the analytical treatment based on the sine-Gordon string theory developed by Marchesoni,[9] the NW and Bordoni peaks are manifestations of the same kink-pair nucleation mechanism, depending on the presence or absence of geometrical kinks/antikinks, respectively. On the contrary, Seeger[10] argued that in most fcc metals two relaxation processes (the NW and Bordoni peaks) occur due to the kink-pair formation in dislocations running along the $\langle 110\rangle$ and $\langle 112\rangle$ directions. In bcc metals the problem is even more complicated due to specific structure of the core of screw dislocations and similar questions exist.

This work provides an additional input to the understanding of the dislocation dynamics in bcc metals by analyzing the internal friction measurements in various iron-based materials with different impurity/defect concentrations. If dislocation pinning due to the presence of geometrical kinks plays a crucial role for the activation of subsidiary peaks in the internal friction spectra, as in the Marchesoni model, the kink-pair activation process should have different behaviors in diluted and concentrated alloys. This could be expected in both fcc and bcc materials since the kink propagation can be strongly modified due to the presence of defects.[11] In Sec. III, the experimental results are presented and discussed. In the same section, a phenomenological model is developed and applied to the internal friction spectra, accounting for the essential experimental observations such as a subtle interplay of the β and γ peaks.

II. EXPERIMENT

The materials used in this study are polycrystalline iron alloys; the nominally pure Fe; Fe-x%Cu and Fe-x%Cr, with a variety of Cu and Cr concentrations; and the Japanese reference quality (JRQ) steel. The details of the sample preparation were published elsewhere.[12–14] All samples are prepared at the temperatures above the miscibility limit, which ensures the attainability of a solid solution at high impurity concentrations. The chemical composition of the investigated materials is given in Table I.

The Fe-Cu samples are prepared using argon arc-melting and zone refinement methods. The resulted ingots were then cold worked after austenisation tempering. To release the stress and to get a well recrystallized material, a final heat treatment at 1075 K for 1 h followed by water quench is performed.[12] After casting, the obtained Fe-Cr ingots were cold worked under protective atmosphere to fabricate plates of 9 mm in thickness. The Fe-Cr model alloys were treated at 1323 K for 1 h in high vacuum for austenisation and stabilization. The tempering was done at 1003 K for 4 h, followed by air cooling.[13] The JRQ steel was produced by BOF-LRF (basic oxygen free, ladle refining furnace) process.[14] After rolling, the plates were heat treated, normalizing at 1173 K, quenching from 607 K, and tempering at 823 K, for 12 h. Finally, the stress relieving was done at 893 K for 40 h.

TABLE I. Nominal composition of investigated alloys.

Material	Nominal composition (wt %)
Pure Fe	(<200 ppm impurities)
Fe-0.1%Cu	0.1% Cu (<30 ppm C)
Fe-0.3%Cu	0.3% Cu (<30 ppm C)
Fe-1.0%Cu	0.95% Cu (~0.1% additional impurities, ~30 ppm C)
Fe-2.5%Cr	2.36% Cr (<200 ppm additional impurities)
Fe-5%Cr	4.62% Cr (<200 ppm additional impurities)
Fe-9%Cr	8.36% Cr (<200 ppm additional impurities)
Fe-12%Cr	11.62% Cr (<200 ppm additional impurities)
JRQ	3.51% impurities (~180 ppm C)

The internal friction measurements are performed in an inverted torsion pendulum[15] operating in free vibration at about 1.8 Hz (with a typical sample size of $1.3\times1.3\times30$ mm^3) in the temperature range between 100 and 600 K. From the free decay signal, the resonance frequency ω and the internal friction coefficient Q^{-1}, proportional to the ratio of the energy dissipated during one cycle to the maximum elastic energy stored in the sample, are determined. The measurements have been performed at the strain amplitude of about 10^{-4} in a He atmosphere with a heating rate of about 1.5 K/min, and no magnetic field is applied. Prior to the measurements all samples are, in a consistent way, subjected to a torsional cyclic plastic deformation by an angle of $\pi/2$ over a length of 30 mm at room temperature. This corresponds to a local deformation of about 6% ($\pi R/L$).

III. RESULTS AND DISCUSSIONS

The internal friction coefficient Q^{-1} as a function of temperature for various cold-worked iron alloys is shown in Fig. 1. All spectra show almost no trace of the Snoek carbon peak (carbon relaxation at 1.8 Hz should be expected at about 310 K) and a weak contribution of the Snoek-Köster relaxation (associated with carbon relaxation at dislocation sites) at about 500 K, which is in agreement with the reported carbon content in these materials. Two main structures, centered at about 205 and 345 K, are clearly observed in the spectra, indicating that, in spite of compositional variety, no drastic microstructural change occurs. Their overall shape, the temperature position of the maximum, full width at half maximum (FWHM), asymmetry, and the background agree well with previous measurements.[16–18] Moreover, both modes completely disappear in well annealed samples which allows their assignation as the β- and γ-relaxation processes. Interestingly, by increasing the impurity concentration, the relative intensity ratio of the two peaks in these alloys changes in favor of the β peak, so that it becomes a dominant feature in the internal friction spectra of Fe-12%Cr (see bottom spectra in Fig. 1). The existence of the β and γ intensity interplay reinforces the argument that these two modes have a common origin. An accompanying effect, ignored further on, is the lowering of the high-temperature background, which

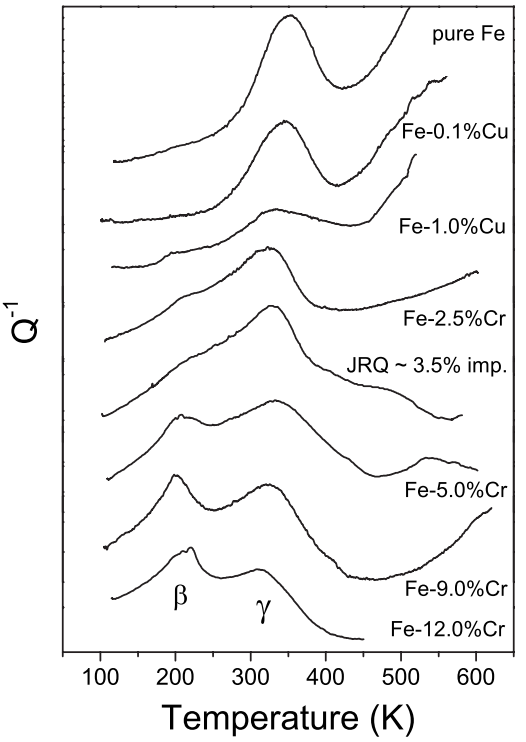

FIG. 1. Internal friction spectra as a function of the temperature of plastically deformed Fe-x%Cu, $x=0$, $x=0.1$, and $x=1$; Fe-x%Cr, $x=2.5$, $x=5$, $x=9$, and $x=12$; and JRQ measured at the frequency of about 1.8 Hz. The same y-axis scale is used for all spectra. The typical background value is about 10^{-3}.

could be understood through the decrease in the ferromagnetic component in the materials and/or the decrease in the lattice fluctuations.[19] In the internal friction spectra of Fe-x%Cu, no trace of the β peak is observed for $x=0$, $x=0.1$, and $x=0.3$ (see Fig. 1), while in the $x=1$ sample the β peak appears as a weak, highly asymmetric structure. At the same time, the γ peak loses its intensity due to the increase in dislocation pinning. It seems that copper is more effective than chromium in reducing the γ-peak intensity. This could originate from stronger Cu-dislocation interaction due to larger local strain field. The strong reduction in the β peak in diluted alloys, as well as in pure Fe (see also Ref. 16), contradicts the interpretation of the β peak as the relaxation associated with the glide of one of the two core structures of screw dislocations, since this process is expected to be active even in the ultrapure materials.[10] Moreover, the β peak is probably not a process analogous to NW relaxation, since the NW peak is observed in pure fcc metals.[20] Moreover, the internal friction spectra show that the increase in the β-peak intensity can be correlated with a decrease in the high-temperature background, which is opposite to Marchesoni's result on pure copper.

Evidently, the experimental observations cannot be explained within existing models, so in the following we focus on the intensity interplay between the β and γ peaks and propose a phenomenological model which accounts for the main observations without pointing to the microscopic origin of the β-relaxation process. We derive a model in which we assume that the number of relaxing kink pairs in a material is

proportional to the probability of such pairs not to be trapped by the impurities, $P=1-l/L$, where l is the average kink-pair size and L is the average impurity-impurity distance. In other words, if a kink pair forms with a preferential (constant) size l (kink-antikink distance), the probability of pair formation depends on the probability of not finding an impurity within a kink-antikink pair. Furthermore, this probability is either proportional to the impurity concentration (c) or inversely proportional to the impurity-impurity distance L. It is further assumed that trapped dislocation segments, $1-P$, contribute to a β-relaxation process whose activation energy is on the order of half the kink-pair nucleation energy. Now, the internal friction coefficient can be calculated by incorporating these probabilities into the relaxation strength of standard equation,[3] generalized to the case of two relaxation mechanisms,

$$Q^{-1} \sim \frac{l}{L}\frac{\omega\tau_{E_1}}{1+(\omega\tau_{E_1})^2} + \left(1-\frac{l}{L}\right)\frac{\omega\tau_{E_2}}{1+(\omega\tau_{E_2})^2}, \qquad (1)$$

where ω is a frequency and $\tau_{E_1}=\tau_0 e^{E_1/k_BT}$ and $\tau_{E_2}=\tau_0 e^{E_2/k_BT}$ are the relaxation times with activation enthalpies (energies) E_1 and E_2, respectively.

The first term of Eq. (1) describes the β contribution (Q_β^{-1}) and the second term the kink-pair contribution (Q_γ^{-1}) to the internal friction coefficient. The fact that dislocation motion is not governed by a single relaxation time and manifested by a large FWHM of the internal friction peaks ($\sim$60 K for the γ peak) is accounted for in the model by incorporating a Gaussian distribution of the relaxation times, $\Phi(\tau)=\frac{1}{g\sqrt{\pi}}e^{-(\tau/g)^2}$. Finally, the normalization of the spectra, $\int\int Q_\beta^{-1}[\Phi(\tau),T]d\tau dT=\int\int Q_\gamma^{-1}[\Phi(\tau),T]d\tau dT$, ensures that there are equal numbers of relaxing events for β and γ processes in the material for $l/L=0.5$. The variation in Gaussian parameter leads to different estimates of the l parameter and normalization condition for l/L, but it does not influence the main conclusions related to the intensity interplay since the functional dependence of I_β/I_γ on impurity concentration is determined via $L(c)$ dependence.

The calculated internal friction spectra, reflecting the change due to the difference in l/L, are shown in Fig. 2. The parameters used in this calculation, $\tau_0=10^{-15}$ s, $\omega=1.8$ Hz, $E_1=0.52$ eV, $E_2=0.96$, and $g_1=8$, $g_2=5$, reproduce well the main characteristics of the spectra, such as temperature positions of the peak maxima and FWHMs. The kink-pair nucleation energy (γ peak) is found to be about $E=0.96$ eV, which is in accordance with previous estimates.[16] By increasing the l/L ratio, the probability of kink-pair formation decreases, leading to the reduction in the γ-peak intensity, in very good agreement with the experiment; see Fig. 1. Simultaneously, the β-peak intensity increases at the expense of the γ peak, since dislocation segments that are not favorable to the kink-pair formation contribute to the β-relaxation process.

Up to now, the choice of l/L was fully arbitrary. This parameter varies between $l/L=0$ and $l/L=1$, which are two extreme cases corresponding to free dislocation motion and fully pinned dislocation segments, respectively. In order to analyze this further, we plot in Fig. 3 the intensity ratio of the

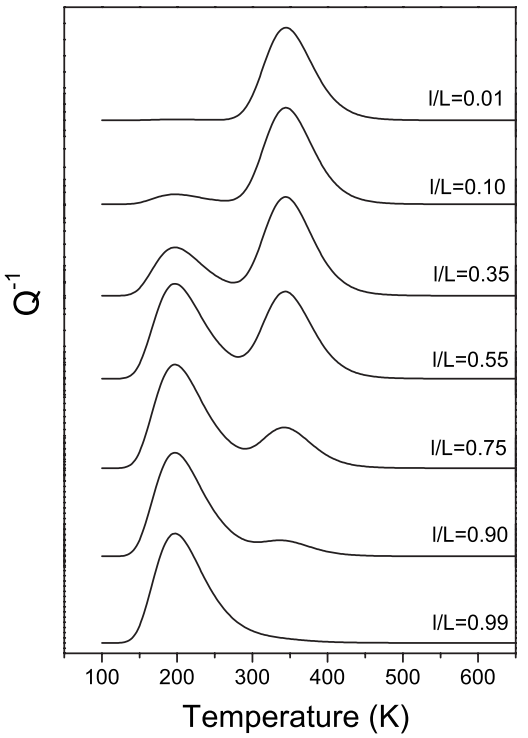

FIG. 2. Internal friction spectra calculated for $l=1.7a_u$ on the basis of Eq. (1) for different l/L values. The spectra are normalized in such a way that there are equal numbers of the single-kink and kink-pair relaxing units in the material for $l/L=0.5$; see text.

β and γ peaks, I_β/I_γ, as a function of the impurity concentration. With increasing impurity concentration, the I_β/I_γ slightly deviates from the linear behavior. Since L describes the average impurity-impurity distance, it depends on the impurity concentration as a linear, square-root, or cubic-root function, depending on whether one considers the dislocation motion to be predominantly one, two, or three dimensional, respectively. In reality the behavior is probably a complex mixture of all three types of motion that might differ in low- and high-concentration alloys. Moreover, it may exhibit the threshold as a consequence of the defect critical size for dislocation pinning. The l may be assumed as independent of

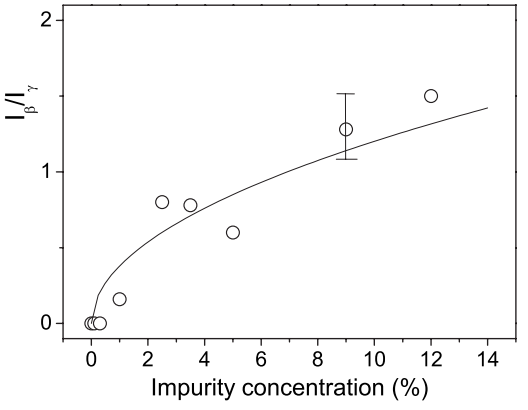

FIG. 3. I_β/I_γ as a function of the impurity concentration. The full line corresponds to the calculated data, and the symbols to the measured data.

the impurity content in the first approximation, since the γ-peak position only slightly changes in all spectra. (Some changes in the peak positions in the internal friction spectra can be traced back to a change in sample size and/or frequency, which produces an uncertainty of about 5%.) We proceed with the most simple scenario, in which there is no clustering in the material and the motion is assumed to be two dimensional (dislocations are mainly gliding in a plane). The intensity ratio I_β/I_γ, shown as a full line in Fig. 3, is obtained from calculated internal friction spectra by taking $L \sim 1/\sqrt{c}$ and $l=1.7a_u$. From the comparison between measured and calculated data. we find that for an impurity concentration around 10% (see also bottom spectra of Figs. 1 and 2), l is on the order of $L/2$, which corresponds to a kink-pair size of about $l \sim 2$ atomic distances (a_u), and that it can be indeed regarded as a characteristic of the material (in this case iron). This is fully in accordance with previous experimental observations showing that the γ-peak position is not significantly affected by the impurities.[3] The estimated l value is somewhat lower than previous findings in the single crystals, giving $l \sim (10-15)a_u$.[21] However, one has to keep in mind that in such concentrated alloys clustering is likely to occur. Thus, the kink-pair size is expected to be at least a factor of 2 higher.[22]

The presence of carbon interstitials might influence the interplay between the β and γ peaks, since Snoek-relaxation energy is in the middle of the β- and γ-activation energies, so a large interaction between carbon and kink pair is expected.[23] Questions related to this issue are the subject of ongoing research.

IV. CONCLUSION

To conclude, we observed the interplay between dislocation-related β- and γ-relaxation peaks in the internal friction spectra of iron-based materials. We argue that the interplay occurs as a consequence of different probabilities for two types of dislocation relaxations in pinned dislocation segments. The model incorporates an increase in the β-relaxation process at the expense of the γ relaxation, giving a square-root type of dependence of I_β/I_γ as a function of the impurity concentration, in a very good agreement with experimental data.

ACKNOWLEDGMENTS

The work was supported by the Electrabel-Suez Tractebel-SCK CEN 2007–2011 convention and by the European PERFECT project.

*mkonstan@sckcen.be

[1] P. G. Bordoni, Ric. Sci. **19**, 856 (1949).
[2] R. Peierls, Proc. Phys. Soc. London **52**, 34 (1940).
[3] A. S. Nowick and B. S. Berry, *Anelastic Relaxation in Crystalline Solids* (Academic, New York, 1972).
[4] A. Seeger and P. Schiller, Acta Metall. **10**, 348 (1962).
[5] C. Domain and G. Monnet, Phys. Rev. Lett. **95**, 215506 (2005).
[6] A. Seeger, Mater. Sci. Eng., A **370**, 50 (2004).
[7] D. H. Niblett and J. Wilks, Philos. Mag. **1**, 415 (1956).
[8] H. Chambers and J. Schultz, Acta Metall. **10**, 466 (1962).
[9] F. Marchesoni, Phys. Rev. Lett. **74**, 2973 (1995).
[10] A. Seeger, Philos. Mag. Lett. **84**, 79 (2004).
[11] G. Gremaud and S. Kustov, Phys. Rev. B **60**, 9353 (1999).
[12] K. Verheyen, M. Jardin, and A. Almazouzi, J. Nucl. Mater. **351**, 209 (2006).
[13] M. Matijašević, E. Lucon, and A. Almazouzi, J. Nucl. Mater. **377**, 101 (2008).
[14] IAEA-TEC.DOC.1230, www-pub.IAEA.ORG/MTCD/publications/PDF/te_1230_prn.pdf
[15] K. Van Ouytsel, A. Fabry, R. De Batist, and R. Schaller, J. Nucl. Mater. **279**, 51 (2000).
[16] H. Mizubayashi, H. Konmüller, and A. Seeger, J. Phys. Colloq. **10**, 309 (1985).
[17] M. Weller and J. Diehl, in *Point Defects and Defect Interactions in Metal*, edited by J.-I. Takamura, M. Doyama, and M. Kiritani (University of Tokyo, Tokyo, 1982), p. 417.
[18] R. De Batist, J. Nucl. Mater. **31**, 307 (1969).
[19] E. Bucher, F. Heiniger, J. Muheim, and J. Muller, Rev. Mod. Phys. **36**, 146 (1964).
[20] G. Fantozzi, C. Esnouf, W. Benoit, and I. G. Ritchie, Prog. Mater. Sci. **27**, 311 (1982).
[21] E. Kuramoto, Y. Aono, and K. Kitajima, Scr. Metall. **13**, 1039 (1979).
[22] If one assumes that during the alloy preparation the two-atom clusters are formed, the impurity-impurity distance is $L \sim 1/\sqrt{c^2}$, thus giving the value $l \sim (4-5)a_u$.
[23] T. T. Lau, C. J. Först, X. Lin, J. D. Gale, S. Yip, and K. J. Van Vliet, Phys. Rev. Lett. **98**, 215501 (2007).