Friction in ferroelastic and martensitic materials

E.K.H. Salje
Department of Earth Sciences
University of Cambridge, Downing street, Cambridge UK

e-mail: es10002@esc.cam.ac.uk

Abstract. Friction during ferroelastic and martensite phase transformations and under external elastic forcing is related to the appearance of jerks as fingerprint for avalanches of moving domain boundaries. It is argued that such avalanches can be observed if the time scale of the experiment is sufficiently long to allow a statistical evaluation and have a time resolution compatible with the characteristic duration of an avalanche. Typical experimental methods include the observation of acoustic emission, resonance elastic spectroscopy, calorimetry, and advanced phonon spectroscopy. The changes of the internal structure of mobile twin boundaries contribute to changes in their mobility and their friction. The local structures of jammed twin boundary patterns are elucidated by computer simulation, their time evolution is similar to that of observed avalanche dynamics.

1. Introduction
The dynamic response of materials measured at high frequencies, such as measured in phonon spectroscopy or resonant Acoustic Spectroscopy, is generally found to be a smooth function of the applied stress in the regime of small stresses [1-4]. These effects have been widely studied in physics and earth sciences over the last 3 decades [5-7]. Non-linear superelastic softening occurs when external stress leads to the movement of interfaces such as twin boundaries or the interface between two phases. Local boundary movements are part of the dynamics of complex patterns with energy dissipation through the emission of phonons. The macroscopic fingerprint of boundary movements, such as a twin boundary displacement, is often the elastic response as seen in the change of stress under changing macroscopic shear of the sample. Note that strain-stress curves are very different for avalanches when the strain is the tuning parameter (~ hard boundary conditions) or whether the stress is applied (~ soft boundary conditions). The real part of the elastic response in either case relates to the elastic moduli while the imaginary part measures dissipation or friction. Previous studies of the dissipated energy revealed Debye type relaxations related to the movement of interfaces such as twin boundaries [8-17]. Jerky elasticity, where the dissipation displays singularities such as spikes, was envisaged theoretically and has been reported experimentally for 3-point bending experiments [18]. In general, for avalanches to occur it is required that thermal fluctuations play a minor role in an a-thermal scenarios and that the system is driven slowly enough in order to ensure that avalanches do.
not overlap. On the other hand, a quantitative study of avalanches requires the use of very sensitive techniques that enable detection of very small transformed fractions characterized on a sub-micrometer length scale and very high time resolution. In the case of structural transitions, including ferroics and multiferroics several model cases were the internal structure of a twin boundary influences the mobility of the boundary were recorded [19-22]. In some cases, other more integral methods including calorimetry have been used to reveal the existence of avalanches [21].

Avalanches have been commonly observed in collapsing porous materials when the applied stress exceeds a small threshold value which allows the first cavities to collapse [23,24]. In additions, the change in the internal structure of twin boundaries also modifies their mobility under elastic forcing [20, 25,26]. In particular, the formation of dipols inside the domain boundaries can lead to pinning and hence may reduce the domain boudnary friction [27].

Figure 1. Friction and elastic moduli in LaAlO₃ and KMnF₃-type compounds. The thermal phase transitions are seen as collapse of the elastic moduli and a steep increase of the friction coefficient in the ferroelastic phase [10,11].

2. Experimental conditions to observe jerks
We argue in this paper that jerks and avalanches are key ingredients for the understanding of the physical processes underlying friction in ferroelastic materials and martensites. Jerks become observable under experimental conditions when the time scale of the experiment is slow enough to distinguish clearly between individual jerks. This condition is not easily met because the integration time per event is often some $10^{-4}$ seconds while the number of events in a largely a-thermal experiment
is related to the total number of particles divided by the number of transforming parts per event. For a sample with $10^{23}$ atoms and on average perhaps $10^{15}$ atoms per avalanche we expect about $10^8$ events for a first order phase transition in ferroelastic materials, such as martensites. If these events were spaced equally in time, we expect the total transformation to happen in $10^4$ seconds. An experiment where a volume transformation is observed as function of the sample temperature requires cooling rates slow enough to detect at least the major events without overlap between them. In this estimate, an appropriate cooling rate for a sample inside the coexistence interval of 10K is 1 milliKelvin per hour, which is an extremely slow cooling rate. Any faster rate would superpose various events and artificially generate the impression of large avalanches, which do not exist in the sample (see Fig. 2).

The avalanche duration is shorter than 1 msec and often reduces to a few phonon times so that the required extremely high time resolution for the measurement of single avalanche profiles is very hard to obtain. Furthermore, the extremely high time resolution has to be maintained over long observation periods to measure a sufficient number of avalanches to determine their statistical distribution. This experimental requirement is hard to match and it becomes clear why so few experimental data can be found in literature.

The situation is somewhat helped if not the full transformation is jerky but where some part involves the continuous propagation of phase fronts through the sample. A typical example is CuZnAl where only 5% of the transformation process involves jerks while the remaining 95% is continuous, Fig 2. Even in this case, the observation of the jerks required heating and cooling rates of some milliKelvin per hour, otherwise signals become corrupted by overlap and cannot be used for meaningful statistical analysis. In summary, good jerk measurements require patience to obtain reliable useful statistical data and a very high time resolution. A typical example for the dissipation by friction in a mechanical shear experiment is shown in Fig. 3.

![Figure 2. Superposition of jerk patterns and smooth background spectra related to the continuous shift of domain structures in CuZnAl [21]](image)
Figure 3 Friction spectrum of CuAlBe during the austenite-martensite transition. The data were measured by a Dynamical Mechanical Analyzer (DMA) at low frequencies. Tan δ is a measure for the dissipated elastic energy [18].

3. The local domain structure and the size dependence of domain boundary friction

The geometrical movement of twin boundaries in ferroelastic materials and martensites has been investigated systematically by computer simulation by Ding and collaborators [28-30]. They have shown that the pattern formation is strongly size dependent (Fig.4) and that the jerk spectrum follows power law distributions only at low temperatures (a-thermal regime). At higher temperatures these authors found that the noise spectrum is best described by a Vogel-Fulcher dynamics with an stretched exponential regime at the thermal crossover [28,29,30]. These simulations clearly identify two basic domain movements (Fig.5) that characterize the friction behavior of ferroelastics: first we encounter large movement of needle domains which become non-reversible each time a needle collapses at another twin boundary or at the sample surface. Renewed needle movement requires the nucleation of a another needle which often involves large nucleation energies. The second movement is the propagation of kinks inside twin walls which are destroyed when the kink reaches the sample surface. The nucleation energy is rather small in this case and new kinks appear readily under strain.
Figure 4. Simulated twin pattern under strain forcing. Note that the complexity of the pattern is constraint to large system sizes [28,29].

Fig. 5 Snapshots of computer simulation of a needle domain (a) and a kink in a twin wall (b).

Under applied external strain the needle domain will progress and retract at the lower end of the needle. Friction is related to the emission of phonons during this movement. A large acoustic emission signal is emitted when the retraction of the needles becomes so large that the needle is destroyed and
the total self energy of the needle is emitted as friction signal (see Fig.3). The kink in (b) moves inside the twin wall under external strain driving. Dissipation is exclusively related to phonon emission until the kink disappears at the sample surface.

In summary, we find that friction in ferroelastics and martensites is related to the movement of twin boundaries and other microstructures. Smooth movements show little friction and can be observed by elastic spectroscopy while sudden, jerky movements involve large friction and are seen as singularities in a multitude of experiments, including calorimetry, AE, and RUS. The experimental observations are well reproduced by computer simulation. The key physical parameter to characterize domain wall friction is the jerk distribution: we find power law distributions at low temperatures and thermally activated Vogel-Fulcher statistics at high temperatures.

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