Final report

for project

OTKA PD131784 – “Adaptive noise clustering during structural changes”

The main purpose of this project was to improve and apply clustering methods for the highly sophisticated evaluation of noise emission signals. With the clustering method, we can make difference between noise avalanches, which were detected during the same measurement, but originated from different noise sources. These sources can be the sample itself and the environment (background noise), as well as different physical phenomena operating in the sample during the same experiment. With the separation of these avalanches we can investigate the structural changes in more detail, and with high accuracy, which are the necessary conditions of the modern noise measurement methods. During the project, I investigated different structural changes with acoustic emission technique. The measurements were carried out with continuous signal recording in order to investigate the properties of the noise events in frequency domain. Using these properties, with the adaptive sequential k-means clustering method, the determination of the active noise sources during the transformation was possible.

Originally, the two structural changes, which were mentioned in the research plan was the martensitic transformation: the investigation of the asymmetry between the forward and reverse phase transformation of shape memory alloys, as well as the ductile-brittle transition of steels. In addition, during the project, I was open-minded to react to the quickly changing interest of the field, and apply our methods to answer actual questions. The main results of the project are summarized as follows:

In the beginning of the first year of the project, I studied the literature. I improved my previous noise evaluating software to implement the clustering method, and I started testing the algorithm on acoustic emission measurements. During the testing I realized, that some modifications are necessary in the literature algorithm for more accurate results. For example, one of the modifications were regarding to the merging of the clusters: it is better to do the merging only after all of the events are classified into a cluster. In the latter case, the clusters contain more events and thus, the uncertainties for the cluster centroids and for the dispersion of the events in the clusters (related to the radius of the cluster) are smaller. Besides this issue, I had to answer several questions, related to the details of the calculation, like what is the best way to calculate the fast Fourier transform of the acoustic emission events with different length, or what is the optimal method to determine the similarity between the power spectral density (PSD) curves of acoustic emission events.

The first experiments were done on a SIM-aged Ni_2MnGa single crystal, which was compared with a not SIM-aged (as grown) one, to study the effect of the SIM-aging on the acoustic emission characteristics of the phase transformation. SIM-aging (stress-induced martensite stabilization aging) is a martensite stabilization heat treatment, which is carried out under uniaxial stress, developed by Yu.I. Chumlyakov and his group. During the work it turned out, that besides the thermodynamical properties of the austenite-martensite transformation (start and finish temperatures, transformation entropy, …) changed, the statistical parameters of acoustic emission also changed: the SIM-aging changed the sign of the asymmetry. Details are in the first publication of the project (László Z. Tóth, et al. "Acoustic Emission Characteristics and Change the Transformation Entropy after Stress-Induced Martensite Stabilization in Shape Memory Ni53Mn25Ga22 Single Crystal." Materials 13.9 (2020): 2174.). This topic was not included originally in the research plan, but it fits very well into the project, and it answered actual questions

The second AE measurement was the plastic deformation of tin at different temperatures (slightly preparing for the ductile-brittle transition of steels, where the main idea was the possible difference between acoustic avalanches during plastic deformation (ductile) as well as cracking (brittle) of the steel). The dominant sources of AE are both the collective motions of dislocations as well as deformation twinning, and the dislocation glide avalanches and intermittent twinning events have different AE characteristics. The deformation rates for this two mechanisms are different at different temperatures; thus the primary deformation mechanism is changing with the temperature. Using classical noise analysing methods, we demonstrated in a publication (Daróczi, L., Elrasasi, T. Y., Arjmandabasi, T., Tóth, L. Z., Veres, B., & Beke, D. L. (2021). Materials, 15(1), 224.), that this difference is visible in the acoustic emission characteristics, and the transition temperate was determined.

For the clustering analysis of the acoustic emission measurements on tin, I had to carry out a new set of measurements, which is more suitable for such an analysis from the technical point of view. There were differences in the processing of the signal coming from the analogue to digital converter, I collected the raw voltage signal, and all the signal processing steps (band-pass filtering, finding the avalanches …) were done offline, after the recording. I made measurements at the following temperatures: -30 °C, 0 °C, 25 °C (room temperature), 50 °C and 80 °C. At temperatures below -30 °C, the system required massive cooling (using liquid nitrogen), which caused high background noise, while at 50 °C and above, the noise level and the number of AE avalanches were too low for a clustering analysis. The analysis of the -30 °C, 0 °C, 25 °C measurements showed, that there are typically 3-4 clusters at each temperature. At a given temperature, all the clusters have significantly different cluster centroids, and after the statistical analysis of the events in the different clusters it turned out, that for example the exponents of the energy distributions are different for different clusters. Moreover, more complex parameters, calculated from correlation between different noise parameters (size-duration, energy-amplitude …) gives different results for different clusters. These findings were published in: **Tóth, L. Z.**, et al. (2022). *Clustering Characterization of Acoustic Emission Signals Belonging to Twinning and Dislocation Slip during Plastic Deformation of Polycrystalline Sn*. *Materials*, *15*(19), 6696.

I had the opportunity to take part in a common research with the Technion – Israel Institue of Technology. In our laboratory, we compressed a single crystalline Ni2MnGa ferromagnetic shape memory alloy in martensitic state to study the structural change caused by twin boundary motion. We used a special equipment to compress the sample with extremely low compression rate, which uses thermal expansion of an alumina rod. We measured the stress, the magnetic and acoustic emission of the sample during the compression. This experiment is unique, because we applied magnetic field in 45° with respect to the axial direction of the sample forms the microstructure, where a twin boundary divides the sample into two regions, each with a single magnetization direction. In this configuration, magnetization changes occur (almost) solely due to twin boundary motion. We found, that there is an approximately linear relationship between the measured magnetic emission (ME) voltage and the nm-scale volumes exhibiting twinning transformation, i.e. there is a definite correlation between ME signals and stress drops, but the magnetic sensor is much more sensitive than the force sensor. Maximum likelihood analysis of statistical distributions of several variables reveals a good fit to power laws truncated by exponential functions.

These findings were published in Advanced Functional Materials: E. Bronstein, László Z. Tóth, L. Daróczi, D. L. Beke, R. Talmon, & D. Shilo, (2021). Tracking Twin Boundary Jerky Motion at Nanometer and Microsecond Scales. Advanced Functional Materials, 2106573. IF: 19.924.

The third year of the project was the year of publications and conferences. I took part in several conferences and with my coauthors, I published several papers. I presented my results in March 2022 on the 16th International Conference on Martensitic Transformations (ICOMAT 2022) with the title of "*Simultaneous Investigation of Acoustic and Magnetic Emission during Jerky Twin Boundary Motion in Single-Crystalline* Ni_2MnGa". This research is in connection with our collaboration with Prof. Doron Shilo and Emil Bronstein from the Technion, Israel. The ICOMAT conference was held online, due to the Covid-19 situation. The next international conference was the Avalanche conference in Debrecen (Hungary) in August, where I gave an oral presentation on "*Scaling of average avalanche shapes for acoustic emission during jerky twin boundary motion in single-crystalline* Ni_2MnGa". Finally, I travelled to Ankara (Turkey) in September to take part on the 12th European Symposium on Martensitic Transformations (ESOMAT 2022), where I gave an oral presentation, entitled "*Acoustic Emission Characteristics and Change the Transformation Entropy after Stress-Induced Martensite Stabilization in Shape Memory* $Ni_{53}Mn_{25}Ga_{22}$ *Single Crystal*".

There was a long-standing question in the crackling noise area, that the energy (E) of a noise avalanche should be proportional to the third power of the amplitude (A_m) of the avalanche, while the area (S) should be proportional to the second power of the amplitude, according to the mean field theory, but in the experiments, they usually observe considerably smaller exponents. This was called as enigma, by Casals et. al. (Casals et al., Sci. Rep. 2021, 11, 5590). Our group offered an explanation for this, namely, that in the theoretically predicted average voltage ($U(t)$) function of an avalanche at fixed size, $U(t) = atexp^{-bt}$ *a* and *b* are not constants, rather *a* has a definite dependence on the amplitude: $a=A_m^{\phi}$. We showed, that this ϕ parameter appears in the scaling relations: $E=A_m^{3-\phi}$, and $S=A_m^{2-\phi}$. Also, besides many other results, we showed, that the rising time (the time has passed since the starting of the avalanche until the time of the maximal voltage Am) can be given as $t_m = A_m^{1-\phi}$. These theoretical findings were tested on *clustered datasets*, and the results were published in: Kamel, S. M., Samy, N. M., **Tóth, L. Z.,** Daróczi, L., & Beke, D. L. (2022). Denouement of the Energy-Amplitude and Size-Amplitude Enigma for Acoustic-Emission Investigations of Materials. *Materials*, *15*(13), 4556.

The focus is increasingly on the average temporal shapes of avalanches, i.e. the average of the detected voltage signal ($U(t)$) of magnetic or acoustic emission signals for a given size (or duration) range of avalanches. Avalanche shapes carry information about the source, if the voltage of the detector is proportional to the average interface velocity, thus the dynamics of transformations can be studied. To obtain a universal scaling function, with which the experimentally determined average shapes for different avalanche sizes agree, we have to rescale the voltage and time axis to have dimensionless units. The most obvious way for this is normalizing the voltage axis with the amplitude and the time axis with the duration. According to the mean field theory, $S \propto A^2$ and $S \propto T^2$, thus both the voltage and the time axis can be normalized by the square root of the avalanche size. It was demonstrated in the literature, that the duration of the avalanche is distorted by the transfer function of the system, while the amplitude and the rising time remained undistorted. Thus, scaling the voltage axis with the amplitude, and scaling the time axis with the rising time seems promising. Moreover, using the results of our paper in Materials (*15*(13), 4556), it is even more beneficial to use the $t_m \propto A_m^{1-\varphi}$ relation and normalize the time axis with $A_m^{1-\varphi}$. I published an experimental study on the average temporal shape of avalanches obtained for acoustic and magnetic emission measurements during jerky twin boundary motion in Ni_2MnGa ferromagnetic shape memory alloy. The twin boundary was moved as the result of slow, uniaxial compression in martensitic state, as it was described in E. Bronstein, László Z. Tóth, L. Daróczi, D. L. Beke, R. Talmon, & D. Shilo, (2021). *Tracking Twin Boundary Jerky Motion at Nanometer and Microsecond Scales*. Advanced Functional Materials, 2106573. The clustering analysis of the results were necessary here as well, to exclude the background vibrations from the evaluation to get more reliable average avalanche shapes. The preliminary results were presented in 2022, on the Avalanche conference in Debrecen, and the final publication is: Tóth, László Z., et al. "Scaling of Average Avalanche Shapes for Acoustic Emission during Jerky Motion of Single Twin Boundary in Single-Crystalline Ni2MnGa." *Materials* 16.5 (2023): 2089.

Regarding to the two examples for structural changes, which were mentioned in the original research plan: I made measurements on several shape memory alloys (both Ni_2MnGa and NiFeGaCo) to investigate the asymmetric behaviour, and during the evaluation, I made clustering analysis. However, in some cases the clusters showed results, which are similar to the preliminary results, the statistical analysis did not show considerable difference between heating and cooling exponents. The possible explanation for the more symmetric behaviour can be that after multiple heating-cooling cycles on the same samples over the years, the sample "learned" the exact way of the transformation. Thus, for the best results, new, freshly produced samples are needed. (Unfortunately, the Adaptamat Ltd, the producer of the Ni_2MnGa samples was closed, and due to the war in Ukraine, we can not purchase NiFeGaCo samples from the Russian Federation anymore, as we did previously.) We started to prepare for the investigation of ductile-brittle transition of steels with the means of acoustic emission. The climate chamber which can be mounted on a tensile test machine is ready, and we fixed a massive Instron tensile test machine which is stiff enough for the experiment. This work needed much more technical developments than expected to carry out precise and reproducible measurements with low background noise. Thus, despite our efforts, this part of the project is not ready, but definitely planned to finish.

Due to the Covid-19 situation, the planned conferences were cancelled, postponed, or were held online. Thus, I could save a significant amount of money, and I could extend the project by 6 months and I could purchase a new computer to replace the old one (which had several problems over the years) in order to securely finish the project. For this, we had to modify the original expense plan.