1. PUBLISHABLE SUMMARY

The main goal of the project was the professional training and career development of the grant beneficiary researcher, Dr. Hovorka, to provide him with the knowledge and tools in the field of computational magnetism focusing on investigating superparamagnetic and hysteretic behaviour of magnetic nano-particle (MNP) systems, and the related thermodynamic aspects. Fulfilling these aims allowed him to tackle the outstanding challenges in developing the MNP-based magnetic energy conversion methodologies with a potential use as sensors in biology, for cancer therapy in medicine, or in magnetocaloric refrigeration, for example. This aim has been fully achieved by enhancing Dr. Hovorka's expertise in fundamental nano-magnetism, by adding complementary knowledge of computational Monte-Carlo, micromagnetic, and atomistic modelling of MNPs, in conjunction with the main objectives of the project originally identified as:

i. Development of advanced multi-scale models of thermodynamic properties of MNPs.
ii. Development of Monte-Carlo modelling approaches to studying interacting complex mixtures of superparamagnetic and blocked MNPs, and identifying the role of interactions.
iii. Exploration of the range of validity and effectiveness of presently used approaches to characterise energy conversion in mixed superparamagnetic and blocked MNP systems.

Generally, understanding the fundamental aspects of heat generation in out-of-equilibrium systems with hysteresis has been challenging due to the fact that general thermodynamic principles governing the entropy production transfer into a system's degrees of freedom are not yet understood. For these reasons, the entropy production (i.e. the non-equilibrium energy loss) generally cannot be computed in a straightforward way, except for cyclic processes in systems subject to periodic driving forces, or for systems close to equilibrium. The objectives have been designed to extend these approaches to non-cyclic processes and to far-from-equilibrium situations, and the main achievements can be summarized as follows.

Development of multi-scale approaches in objective (i) as a part of training strategy lead to important developments with Dr. Hovorka's involvement, providing explanations for intriguing phenomena during ultrafast magnetization processes in complex materials, with potential applications in the future generation all-optical recording technologies. This work is currently under review in the high profile peer reviewed journal **Scientific Reports (Nature)**.

In accordance with the objectives (ii), one of the major achievements was the development of kinetic Monte-Carlo methods and a semi-analytical master equation approach of the Markov process theory with applications to evaluating the energy loss generation during the magnetization decay process starting from a hysteretic state and proceeding towards equilibrium. The approach allowed identifying the relaxation time spectra, which needs to be accounted for to fully quantify the energy loss behaviour.

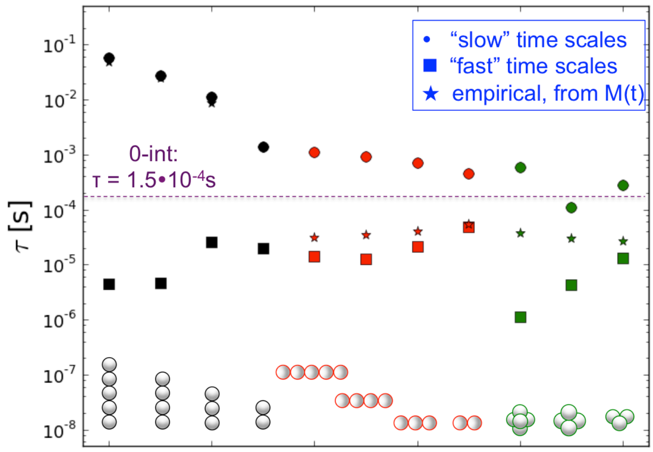

Figure 1 – The shortest (squares) and longest (circles) natural timescales associated with chains and clusters of interacting magnetic nanoparticles as shown in the figure. For comparison, the mean timescale directly obtainable from the magnetization decay measurements is added (stars), and the non-interacting case time constant (dashed line).

Figure 1 shows the differences of the range between the shortest and longest natural timescales in various geometrical arrangements of MNPs, such as chains and clusters, which turns out to be as large as several decades. Thus a purely superparamagnetic heating is expected for external field frequencies slower than the structure's longest natural timescale, while hysteretic heating is expected for frequencies higher than the fastest timescale. For external field frequencies within the natural timescale distribution, both contributions to heat generation need to be accounted for by superposition. It is worthwhile mentioning that these differences are entirely due to the dipolar interactions, whose directional character brings into play the dependence on the details of geometry of arrangement of particles within clusters. These results are significant and have been presented in a number of international conferences such as the APS March Meeting 2012, 2013, HMM 2013 conference, and MMM and Intermag conferences. A high profile publication is currently under consideration for publication in **Physical Review Letters**.

The aims of the objective (iii) to test the fundamental thermodynamic relations and empirical formulas for analysis of nanoparticle systems, led to a number of interesting applications. On one hand is a proposal for new (giant) magneto-caloric type materials with a vast potential for applications in magnetic refrigeration, which resulted in a high profile publication involving Dr. Hovorka in **Nature Materials 12, 52 (2013)**. The scaling relations to be used to relate the hysteresis loop properties and the energy losses at different measurement frequencies, have been derived in the the paper **Applied Physics Letters 101, 182405 (2012)**, in close collaboration with the Seagate Inc., Fremont, CA, USA, the major world leading hard disk drive manufacturer. Dr. Hovorka was invited for a research stay in the Summer 2013 to help implement this work. The quantitative analysis of atomistic modelling data for future high performance magnetic recording has been studied, leading to a publication in **Applied Physics Letters 101, 052406 (2012)**, again with a close involvement of researchers from Seagate Inc. A number of additional publications are anticipated in the near future, as listed under the disseminations activities section of the report.

Thus Dr. Hovorka's training has not only been fully concluded but the overall success of the project leads to anticipation that the outcome of the research work will facilitate developing new sensing and heat generating applications in biomedicine. The drive for advancing the MNP based hyperthermia cancer treatment methodology, for example, is a worldwide competition, with major players in the US and Asia, Spain, and Germany. In this way, the present work substantially contributes to Europe's excellence and competitiveness in these areas of research and in the field of applied nano-magnetism in general. The key is also the knowledge transfer to the host institution, The University of York, which adds to maintaining its worldwide leadership in the area of computational magnetism towards the new fundamental aspect advances and also on the industrial scale. Possible commercial joint ventures or spin offs based upon the present research may result in a direct economic benefit and return to the European Union. It also adds to the long-term scientific prospect for Dr. Hovorka himself, as he was able, during his fellowship, to build a solid scientific base to pursue his independent career beyond the time frame of the current fellowship. He plans to contribute to this fundamental level of research in his new post as lecturer in the University of Southampton.

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