Postdoc Fellowships for non-EU researchers

Final Report

Name	Dr. Anton V. DOLGIKH
Selection	2011 Coopération S&T internationale
Host institution	Université Libre de Bruxelles
Supervisor	Prof. Daniel S. KOSOV
Period covered by this report	from 15/01/2012 to 15/07/2013 (18 months)
Title	Heat conductivity of lattices with dipole interaction (Nonequilibrium statistical mechanics, theoretical physics)

1. Objectives of the Fellowship

The main objective of the project has been the determination of heat conductivity in lattice systems of particles with dipole interaction and the study of the different parameters controlling heat conductivity. Heat conductivity is one of the main irreversible processes through which energy is dissipated in natural and artificial systems. This uncontrolled dissipation is part of natural phenomena, but it is a major preoccupation in modern technology confronted to limited energy resources. Although known for long, the fundamental physico-chemical mechanisms responsible for energy dissipation are still sparsely known and their knowledge remains nowadays more qualitative than quantitative. Fossil fuel has been slowly accumulated on Earth during hundreds of millions of years and is nowadays consumed at a high rate. Even if energy is conserved by the first law of thermodynamics, the problem comes from the second law of thermodynamics, according to which entropy is produced in an irreversible way, except at equilibrium where every macroscopic flux is ended forever. On Earth, heat is dissipated into the environment and radiated to the dark sky at night, pure substances of all sorts are consumed and mixed together in irreversible ways. Recycling costs energy that is dissipated irreversibly. The more we recycle, the more we consume and dissipate energy. Yet, little is known on the irregular motions of atoms and molecules in matter at the origin of energy dissipation. The aim of the present project is to shed some light on the microscopic mechanisms of energy dissipation by heat transport in systems of particles interacting by their electric or magnetic dipole. Contrary to the Coulomb interaction which has a very long range between electric charge, the dipolar interaction has a smaller range, but still longer than short-ranged interactions such as the van der Waals forces between neutral atoms or molecules. The dipolar interaction exists in crystalline magnetic solids, as well as in magnetic liquids such as ferrofluids, in magnetic gases, or at corresponding solid surfaces. The dipolar interaction is also important in water confined in quasi one-dimensional carbon nanotubes, which is of great interest for nanosciences and nanotechnology. By the dipolar interaction, energy can be transported from particle to particle through the system forming an ordered or disordered lattice. This energy is in the form of heat and it is flowing from high to low temperature regions, hence energy dissipation.

2. Methodology in a nutshell

The project concerns nonequilibrium statistical mechanics, which is part of theoretical physics. The advent of fast computers is allowing us to simulate from first principles the motion of the interacting particles and their dipole according to their known microscopic Hamiltonian mechanics, supposed to follow the classical rules. This motion is ruled by ordinary differential equations for the angles the dipoles take with respect to a reference frame. The simulations of this motion allow us to compute the statistical properties responsible for heat transport and energy dissipation. In particular, the temperature profile can be obtained, as well as the heat conductivity for different disorder strengths. Moreover, spatial correlations between different dipoles can also be calculated, which is essential to understand the nature of the heat flow under different nonequilibrium conditions. Besides, analytical approaches can be used in order to get mathematical expressions for the spatiotemporal correlation functions in the nonequilibrium steady state. The comparison between the numerical simulations and the analytical approaches allows us to determine the key properties and parameters controlling heat conductivity and thus energy dissipation in the system of interest.

3. Results

The understanding of out-of-equilibrium low-dimensional systems has been a challenging problem for decades. This topic covers a large variety of important problems of modern physics concerning, for example, the necessary conditions for the observation of the Fourier law; how to achieve and manipulate directed transport in systems with Brownian motion; how to gain a useful work in nonequilibrium; how to control the energy transport in one- and two-dimensional assemblies of large organic molecules with high dipole moment arranged on a surface; the mechanism of the transition to chaos in nonlinear chains; the necessary conditions for the occurrence and existence of the temporally periodic and spatially localized excitations in nonlinear chains; the unicity of the nonequilibrium steady state in nonlinear chains; among others problems. Nonequilibrium processes in low-dimensional systems are also of practical and technological interest because of the recent advances in nanofabrication. In this work, we consider an out-of-equilibrium one-dimensional chain of particles interacting with each other via the classical dipolar potential. Our interest is twofold. First, we consider a heat conduction in such chains. Second, we study the emergence of new correlations in the system caused by a heat flow.

We approximate a particle by a point dipole placed at a fixed position on a line. This approximation is valid for different real physical systems. For instance, molecules of artificial molecular rotors contain one or several chemical groups with substantial dipole moment. While the rest of the molecule is kept fixed on a surface, these groups rotate. The dipole chain model is also intimately connected with ferrofluids or solid-state magnetic dipoles. The point dipole approximation is valid for a single-file water chain. The water in narrow single-wall carbon nanotubes forms a strongly ordered one-dimensional chain. Each water molecule is connected by two hydrogen bonds to neighbor molecules, instead of four bonds in bulk, and also interacts with the carbon atoms of the nanotube. The latter interaction is weak compared to the dipolar one, but owing to the high density of the carbon atoms, it is not negligible and leads to the additional stabilization of the water in nanotube. The hydrogen bonds in 1D water are energetically stronger and possess longer lifetime than ones in a bulk. Both of these facts lead to the formation of the stable water molecules chain in narrow carbon nanotubes or pores. Moreover, the equations of motion of dipole chain can be treated as a particular case of Kuramoto model. In turn, this model describes in a broad sense the synchronization in dynamical system of coupled oscillators. It is clear that this formulation is related to a number of phenomena from synchronization of cells in human organism to phase transitions or Brownian motors. The systems with dipole-dipole interaction occur not only in a classical physics but also play an important role in quantum world, for instance, in the rapidly growing area of cold dipolar atomic gases. For example, in the high density limit, single-file quantum dipoles can also form a lattice with a strong localization of atoms near lattice sites.

By carrying out the numerical and analytical analysis of a classical chain of particles with dipole interaction, we have studied in detail the properties of heat transport in ordered dipole chains, as well as in disordered chains in order to determine the effects of disorder on heat transport. As we shall see, these effects are drastic. Moreover, we have shown the existence of long-ranged spatial correlations induced by the heat flow under certain conditions.

I. Analytical results

A. Nonequilibrium dynamics

A chain of dipoles has a microscopic dynamics supposed to be ruled by a classical Hamiltonian function. The dipoles interact over a range limited by a cutoff radius N_{cut} defined as the number of dipoles in the range of interaction. Moreover, the dipoles at both ends of the chain interact with heat reservoirs at different temperatures through Langevin fluctuating forces. The heat current from one reservoir to the second has an expression that is deduced from the Hamiltonian function. By The system is linearized and, by Fourier analysis, the average value of the heat current can be decomposed into frequency components. The positions of the dipoles play an essential role. They can be regularly distributed in an ordered chain, or randomly distributed with a spacing taken according to a Gaussian distribution to model a disordered chain. The variance of this Gaussian distribution characterizes the strength of the disorder.

B. Correlation functions in nonequilibrium steady state

Mathematical expressions are obtained for the time correlation functions of the dipoles. These correlation functions are decaying in time over a characteristic time that determines the relaxation time of the dipole fluctuations. Here also, the time correlation functions can be decomposed into frequency components. The longest relaxation time corresponds to the frequency with the smallest absolute value of its imaginary part, which provides a criterion to characterize the role of relaxation in heat transport.

II. Numerical results

A. Heat conductivity and temperature profile for disordered chains

First, we have established that ordered dipole chains support ballistic transport and their behavior is similar to the one of harmonic lattices, e.g., there is no temperature gradient. However, ballistic regime can be destroyed by disorder in the position of dipoles. We have used the aforedescribed model where the positions of dipoles are taken as Gaussian random variables. The dispersion of the distribution plays the role of the disorder strength. This model allows us to calculate the temperature profile and the heat conductivity of the disordered chain using numerical simulations.

The quantitative measure of heat transport in media is the heat conductivity. To compute this coefficient, we first need to find the heat current, which is given thanks to Hamiltonian mechanics. The calculation of heat current is straightforward for ordered dipole chains. In the case of disordered chains, we have to average the heat current over 200 realizations of the dipole positions. The obtained dependence of the heat conductivity on the chain length shows the transition from ballistic transport with infinite heat conductivity (for infinite chains) to diffusive transport with finite heat conductivity, and eventually the high level of disorder results into thermal insulating state. For ordered chains, the heat conductivity is proportional to the length of the chain. For disordered ones, the conductivity deviates from the linear dependence. The observed change of transport regimes is an example of a very general conductor-insulator transition induced by disorder. The question of interest is whether ballistic transport takes place as a result of the linearization of the original equations of motion, or if it is an intrinsic property of the model. To answer this question, we numerically integrated nonlinear stochastic dynamical equation of motion with recently developed Langevin dynamics integrator. The time step in the numerical integrator is 0.003 MD units. First, we wait for 10^8 time steps to bring the system to the nonequilibrium steady state, then we perform the production run for additional 10^7 time steps and computed the average heat flow. The results show that linearized model gives a very good approximation for the exact heat conductivity. This observation convincingly demonstrates the applicability of the linear approximation for the set of model's parameters used in our study. The disorder also affects the temperature profile of the chain. To show this result, we have calculated the temperature profile for different values of the disorder strength. We use the kinetic definition of local temperature. For ordered chains, the temperature profile coincides with the one in harmonic chains; i.e., the temperature of the internal dipoles is the average of the temperatures of thermostats, and the temperatures of the leftmost and rightmost dipoles are equal to the temperatures of the corresponding thermostats. Disorder destroys the flat temperature profile and generates the formation of temperature gradient. The size of the region of the chain, where it occurs, depends on the disorder strength. Such behavior of the temperature profile could be caused by the localization of the elementary excitations under the influence of disorder and, in this respect, it resembles the well-known Anderson localization transition. The fundamental properties of heat conduction have been recently considered in quantum wires in relation with Fourier's law. Under quite general physical assumptions, an analytical expression can be obtained for the temperature profile with a smooth step over a characteristic length that decreases as the disorder strength increases. The case of the ordered chain corresponds to an infinite value for the characteristic length. For disordered chains, the characteristic length takes small values compared to the system size, and one observes a steep temperature gradient near the center of a chain, while the parts of the chain close to the edges are thermalized at the temperatures of the corresponding thermostats.

B. Nonequilibrium correlation functions and relaxation time to the steady state

We have estimated the dipole relaxation times for different values of dipole moments. The results demonstrate that the relaxation time decreases as the dipole moment increases. Indeed, the time of the relaxation toward a nonequilibrium steady state is determined by the energy transfer inside the system. A higher magnitude of dipole moments results into a more effective energy transfer between dipoles and, consequently, the relaxation to the steady state runs faster. Two main factors influence the relaxation time in the dipole chain. The first one is that the interaction strength between the dipoles results into the decrease of the relaxation time, and the second one is that the chain length results into the increase of the relaxation time.

Furthermore, the space correlations in the chain have been studied by calculating the spatial correlation function between the dipole orientations all along the chain. These spatial correlation functions are defined by averaging over many realizations in order to display the smooth behavior of space correlations. Remarkably, we have observed the emergence of new long-range correlations in nonequilibrium steady states, which are not present in thermal equilibrium. Between the equilibrium and nonequilibrium states, the spatial correlation function can take values differing by several orders of magnitude for dipoles lying at a distance of five and more lattice sites away from some fixed dipole. The equilibrium correlation function decays fast while the nonequilibrium one weakly varies on the length scale of the chain.

One more noteworthy peculiarity of space correlations is observed. Altering the range of dipole-dipole interaction by changing the cutoff radius N_{cut} of the interaction range strongly affects the space correlations in the system. Numerical evidence shows that the magnitude of the correlations is substantially different for chains with $N_{\text{cut}} = 2$-4 or N_{cut} larger than 7. For N_{cut} larger than 7, there is a long-range order in nonequilibrium states that disappears in equilibrium states. This long-range character of the space correlation function is due to the heat flux and the behavior of the equilibrium correlation function supports this conjecture. We have published convincing numerical evidence demonstrating that, without heat flux, i.e., in an equilibrium state, the space correlation function decays faster with the distance than in a nonequilibrium state. The link between the spatial correlation function and the heat flux originates from the fact that the coupling to the thermostats results into the interaction between different eigenmodes of the chain. Indeed, the space correlations are becoming long ranged as the coupling of the chain to the thermostats increases.

Finally, we have considered the influence of disorder on space correlations. Surprisingly, the effect of the disorder turns out to be different from strictly destructive although the long-ranged correlations continue to manifest themselves under nonequilibrium conditions. For a small amount of disorder, the space correlations are stronger than in an ordered chain, whereas for higher values of the disorder strength they become weaker. To quantitatively estimate this result, we have introduced the correlation length ζ defined as the minimal number of sites n_ζ such that the dipole-dipole correlation function is lower than 10^{-3} in absolute value. This correlation length increases as the disorder is switched on and, thereafter, decreasing as the disorder strength increases. This result is very intriguing because from general considerations disorder should break the long-range order in a system. Moreover, it is important to notice that the enhancement of correlations is almost independent of the correlation range. The correlation functions calculated with $N_{\text{cut}} = 2$ and $N_{\text{cut}} = 9$ are close enough to say that the effect of disorder weakly depends on N_{cut}. Another important point is the absence of this effect in equilibrium conditions. To elucidate the origin of this amplification of long-range correlations by disorder, we have compute the local temperature profile along the dipole chain. In the ordered case, the temperature is flat along the chain and changes only for the dipoles at the interfaces. If we introduce a small disorder, a linear temperature gradient develops along the entire chain, which couples all dipoles and leads to the increase of the long-range correlations. A further increase of the disorder results into a strong localization of the thermal gradient in the middle of the chain. Part of the dipoles become « uncoupled » and we observe a weakening of the correlations, as expected in this regime where the chain becomes insulating.

III. Summary

In the present work, we have considered a chain of one-dimensional dipole moments connected to two thermal baths with different temperatures. The system is in a nonequilibrium steady state and heat flows through it. Assuming that the fluctuations of the dipole moment are small, we have developed an analytically solvable model for the problem. The effect of disorder is introduced by randomizing the positions of the dipole moments. We have shown that the disorder leads to Anderson-like transition from conducting to a thermal insulating state of the chain. It is also shown that the considered chain supports both ballistic and diffusive heat transports depending on the strength of the disorder. We have demonstrated that nonequilibrium constraints lead to the emergence of long-range correlations between dipoles along the chain, which suggests that the interplay between nonequilibrium and next-to-nearest-neighbor interactions results into the emergence of long-range correlations in low-dimensional classical systems.

To obtain these results, we have carried out a numerical and analytical study of the classical dipolar chain under out-of-equilibrium conditions. We have approximated the nonequilibrium dynamics of the chain by a system of linearized stochastic differential equations. Accordingly, all the quantities of interest were expressed solely through the elements of the one matrix capturing the linearization. We have focused on two basic aspects of the chain: heat conduction and correlations. To study the heat conduction, we derived a closed expression for the heat current. In this way, we have established that ordered dipole chain supports ballistic transport and their properties resemble ones of the harmonic lattice, e.g., the heat current is proportional to the difference of temperatures in the thermostats and not to the temperature gradient. This fact points to the violation of Fourier's law in the ordered dipolar chain. Nevertheless, ballistic transport regime is destroyed by a disorder introduced with a random distribution of the dipole positions. We used the simple model where positions of dipoles are imposed to be Gaussian random variables. The dispersion of the distribution plays the role of disorder strength. Within the adopted model of disorder, we calculated the temperature profile and heat conductivity. It has been observed that heat conduction undergoes the transition from ballistic to diffusive transport. In the diffusive regime, the heat conductivity decreases as we increase the chain length. The diffusive regime is also characterized by establishing a temperature gradient in chain. The deformations of the temperature profile in disordered chains have been shown to be consistent with recent results on the derivation of Fourier's law in quantum wires. Similarly to the quantum case, there are two different length scales in the present problem of heat conduction. The first one corresponds to the localization length. The second one corresponds to the thermal length. These two characteristic lengths can be very different. In the model considered in the present work, the estimation of the localization length is complicated by the presence of the strong correlations between the eigenmodes. This question is left for further consideration. We have constructed the exact formula for dipole-dipole correlation functions. This allowed us to estimate the relaxation times and to reveal the slowing down of the orientation relaxation as the system size increases. The situation with spatial correlations is more subtle because they are affected by different factors such as the temperature difference between the thermostats, the dipole moments, and the interaction range (cutoff radius N_{cut}). The most prominent feature of the nonequilibrium state is the emergence of the long-range correlations for $N_{\mathrm{cut}} > 7$. It is especially important because usually the model with only nearest-neighbor interaction is considered in the majority of work about out-of-equilibrium low-dimensional systems. It is worthwhile to notice that the value of cutoff changes the heat conductivity only slightly.

In conclusion, we have clearly demonstrated that the long-range behavior of the correlations is caused by the combination of two factors: heat flow and the relatively long range of dipolar interactions. The nonequilibrium drive is essential because it leads to the coupling between the heat-carrying modes of the systems and the interaction between these eigenmodes is necessary for the emergence of long-range structures. One can generalize this conclusion by formulating the conjecture that the emergence of long-range correlations in a one-dimensional system is possible under nonequilibrium conditions when next-nearest-neighbor interactions are included.

4. Perspectives for future collaboration between units

The study carried out in the present project is raising scientific questions and opening new perspectives for future investigations. An important question is to understand how the localization length is affected by the presence of strong correlations between the eigenmodes. In this regard, an open issue is the study of the role of nonlinear excitations, such as discrete breathers, in the process of energy transfer in chains with dipole interaction. A further open problem is to consider a lattice of interacting dipoles evolving in time by quantum dynamics.

Dr. Anton V. Dolgikh is now returned at the Mathematical Physics Department of Voronezh State University (Voronezh, Russia) and we hope to continue the collaboration on these theoretical issues of nonequilibrium physics. Besides the Physics Department of the Université Libre de Bruxelles (Brussels, Belgium), the collaboration would now also include the School of Engineering and Physical Sciences of the James Cook University (Townsville, Australia) where Daniel S. Kosov is Associate Professor.

Beyond the expansion of knowledge with the advances in nonequilibrium statistical mechanics that the present project has allowed, such a collaboration is promising to solve the aforementioned open questions in the study of heat transport and energy dissipation in dipole systems and should lead to international cooperation between the different research units in this field of science.

5. Valorisation/Diffusion (including Publications, Conferences, Seminars, Missions abroad...)

The results of this research have been published in the prestigious journal « Physical Review E » of the American Physical Society, which has a broad international readership following:

Anton V. Dolgikh and Daniel S. Kosov,
Out-of-equilibrium one-dimensional disordered dipole chain,
Physical Review E **88**, 012118 (2013) pp. 1-11 [DOI: 10.1103/PhysRevE.88.012118] ;
preprint arXiv :1304.5060

6. Skills/Added value transferred to home institution abroad

The objectives of the project has been reached and the results have been published in the article cited here above in one of the major journals of the field.

Besides, the postdoctoral visit of Dr. Anton V. Dolgikh in the research unit of Professor Daniel S. Kosov provides an important training in the field of nonequilibrium physics and transport theory. Not only the study and the publication of the article offer a significant promotion, but the postdoctoral visit has also provided the opportunity of learning about the different areas of the field thanks to the implication of the hosting unit in several projects of the physics of complex systems and statistical mechanics at the frontier between physics, surface science, chemistry, and biology.