

Sharing the same dream ¹

Department of Physics, Nagoya University

Ichiro Terasaki²

1 The first contact

Before talking about the memory with Tasaki sensei — in this memoir, I will respectfully call him so as we did while alive— I would like to introduce myself. I am an experimentalist in the field of solid state physics, mainly studying the transport properties of strongly correlated electron systems realized in transition metal oxides and organic conductors. I am interested in unconventional properties arising from strong correlation, and have searched for new materials exhibiting new properties and/or functions that have never been discovered before.

Originally, my first love started from high-temperature superconductivity, when I was a graduate student in Shoji Tanaka Laboratory, The University of Tokyo, where the high-temperature superconductivity was first verified after the historical discovery by Bednorz and Müller in 1986.[1] At that time, I was a witness who saw that the world record of the transition temperature was updated in the susceptometer in our laboratory. This impact was big enough to change my life, and I was involved in the study of high-temperature superconductivity of copper oxides since then. But soon I was bored with too-detailed and nitpicky studies on their superconducting properties, feeling that everyone seemed to forget the initial excitement over the superconductivity above the liquid nitrogen temperature. I had been hungry for new, unprecedented and exciting physics arising from strongly correlated electrons that was believed to be the background of the high temperature superconductivity. For this reason, I extended my research area to the transition-metal oxides related to the high-temperature superconducting oxides. It was fortunate for me to find the spin-Peierls transition in CuGeO_3 [2] and the high thermoelectric conversion efficiency in Na_xCoO_2 , [3] by the time when I got a tenure position at Waseda University in 1997.

It was a job interview for a new professor in our department in the summer in 1999, when I first saw Tasaki sensei. He presented his representative work on irreversibility from reversible dynamics using extended baker's transformation. I was shocked by his research attitude and his

¹This is a manuscript for the memorial compilation on Professor Shuichi Tasaki

²E-mail: terra@cc.nagoya-u.ac.jp



Figure 1: The cover page of the paperback for the public. The title can be read as “Frontiers of Applied Physics.” Tasaki sensei and myself were the co-editors and co-authors of the book.

way of thinking, and thought how smart he was. Although this impression should be somewhat discounted because an unfamiliar research field always seems attractive to non-specialists, it is true that I became his “big fan” since then.

2 The first collaboration

Tasaki sensei was very active not only for his research but also for managerial works in our department. At that time, we were often asked what was the uniqueness of the Department of Applied Physics that differentiated it from the Department of Physics in Waseda University. In order to accommodate students as good as possible, the originality of the department is being more and more important recently, particularly in the case of private universities like ours. Thus we tried various things to establish the identity of our department. Some of them were to make a booklet for our department and to write a paperback for the public. The cover of the paperback is shown in Fig. 1, in which Tasaki sensei and myself were the co-editors and co-authors. Through such works, we began to exchange our idea about physics.

Fortunately my work on Na_xCoO_2 attracted considerable interest. Everytime I was invited to a conference, I had to think deeply of the nature of the thermoelectrics. In my personal view, a central issue on thermoelectrics should be focused on how much heat one electron can carry. I like this idea; heat is a macroscopic quantity while electron is a microscopic object. In this

"entropy production"

$$\Sigma_t = \frac{1}{k_B T} \int_0^t ds v_0 \cdot f_{\text{trap}}(s)$$

$$f_{\text{trap}}(s) = -k(x(s) - x_0(s))$$

$$\therefore \Sigma_t = -\frac{k v_0}{k_B T} \int_0^t ds [x(s) - v_0 s] = -\frac{k v_0}{k_B T} \int_0^t ds z(s)$$

$$z(s) \triangleq x(s) - v_0 s = x(0) e^{-\frac{k}{\gamma} s} - \frac{\gamma v_0}{k} (1 - e^{-\frac{k}{\gamma} s}) + \frac{1}{\gamma} \int_0^s ds' e^{-\frac{k}{\gamma}(s-s')} f(s')$$

$$\langle z(s) \rangle = \langle [x(s) - v_0 s] \rangle = -\frac{\gamma v_0}{k} [1 - e^{-\frac{k}{\gamma} s}]$$

$$x(s) - v_0 s = \langle x(s) - v_0 s \rangle = x(s) - \langle x(s) \rangle$$

Figure 2: Tasaki sensei's hand writing. This calculation was done to explain Wang's experiment published in 2002.[4] A phrase of "entropy production" is seen at the top.

respect the thermoelectrics is a bridge between micro and macro, which needs development of thermo-statistical physics in non-equilibrium states. I often discussed such issues with Tasaki sensei, and I gradually got interested in unsolved problems on steady-state thermo-statistical physics. In fact, I was much influenced by him.

In 2002, I found a paper entitled "The experimental demonstration of violations of the second law of thermodynamics for small systems and short time scales" by Wang et al.,[4] and asked Tasaki sensei's opinion. He seemed to get interested in this paper as well, and in a few days, he showed me a calculation to the above paper, which cast some doubt on their conclusions. Figure 2 shows a part of his hand-writing, where a phase of "entropy production" is seen at the top.

We decided to submit a comment to Wang's paper, which is still now available in the cond-mat archive.[5] In the comment, we claimed that their finding was explained just in terms of random motion due to the random distribution of the initial position of a particle. As expected, we received a harsh counterargument from the authors. Tasaki sensei was so tenacious that he replied to their criticisms, but unfortunately after several exchanges, we failed in publishing our comment. Nevertheless I thought that this experience was really valuable to me, for I recognized that Tasaki sensei and myself were being confronting similar issues.

3 Giant nonlinear conduction

It was the end of 2003 that we discovered giant nonlinear conduction in the organic conductor θ -(BEDT-TTF)₂CsZn(SCN)₄. [6] In this particular material, the charge ordered domains with different patterns coexist and are randomly frozen at low temperature, forming a glassy state of charge ordering. The charge order is an ordered state in the strongly correlated electrons where electrons are localized at every two sites owing to the long-range Coulomb repulsion. In our organic conductor, the charge ordered domains are highly disordered and “weakened”, and consequently are able to be controlled by external currents. Suppose the charge order as a kind of electrons’ ice. Then you can understand that the high resistant state at low temperature of this organic conductor is simply due to the fact that electrons are in the solid state, where all the electrons are no longer mobile. If this ice are melted by external currents, then the conductivity will increase dramatically by increasing the number of free carriers. This is indeed what we observed, which was verified by the x-ray diffraction in electric fields. [7] I was excited to see this phenomenon, and wondered if this was truly a non-equilibrium phenomenon in strongly correlated electrons.

After this nonlinear conduction was first measured by a graduate student, Koichi Inagaki, I talked this to Tasaki sensei at the master thesis defense in February 2004. Surprisingly, he studied a similar subject independently, and had a preliminary result for non-equilibrium Peierls transition, where he found that the external voltage suppressed the Peierls order parameter. After intensive discussion, he decided to calculate the voltage-current characteristics. Dr. Shigeru Ajisaka, who is one of the organizers of this special volume —here I will call him “Ajisaka kun” by expressing my friendship to him—, did the calculation, and obtained the results which were almost identical to the experimental data we observed.

In April 2004, an undergraduate student, Fumiaki Sawano, discovered that the nonlinear conduction was a single-valued function of current, while a multi-valued function of voltage. Tasaki sensei and Ajisaka kun studied the current dependence of the order parameter, and found that the order parameter was certainly single-valued as a function of current. We got excited and strongly felt that we were going to the right direction. Sawano and myself fortunately succeeded in publishing this work in *Nature*, and the nonlinear conduction in strongly correlated systems have received considerable attention.

In spite of the initial success, it took four years to summarize the theoretical work for the non-equilibrium Peierls transition. In the course of this, we tried to extend this theory to the charge ordered state or the Mott insulating state, but such trials were not successful. The final paper of 30 pages was written by Ajisaka kun and Tasaki sensei, and I am deeply honored to be one of the coauthors. [8] Through the completion of this paper, I noticed that the gap equation found by

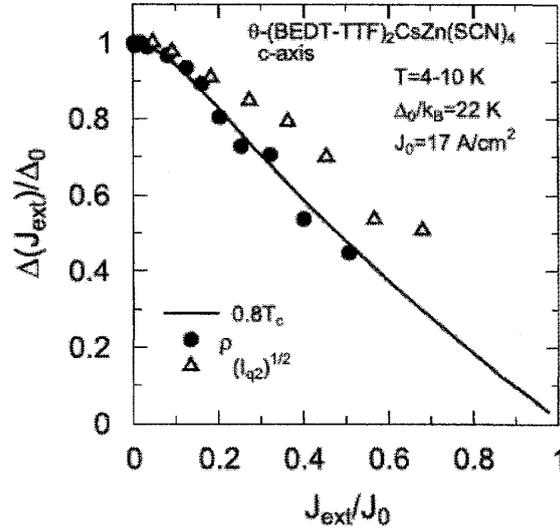


Figure 3: The comparison the experiment done by my group and with the calculation by Ajisaka and Tasaki. The solid curve is the calculation with a reasonable value of $T/T_c=0.8$. The circles and triangles are the experimental data from the resistivity and the x-ray diffraction, respectively. The experimental data are properly normalized with the parameters of a critical current J_0 and an energy gap Δ/k_B at 0 K, which are written in the right upper part of the figure.

Ajisaka and Tasaki was very similar to the gap equation of the non-equilibrium superconductivity by Owen and Scalapino.[9] I remember that all of us were much excited to see that, and felt that “true” physics included something universal.

We compared the calculation with the experiment, and found that the consistency was reasonably good. At last, experiment meets theory; Figure 3 shows the energy gap plotted as a function of external current.[10, 11] The gap roughly linearly decreases with current, which can be understood in terms of excess injection of quasi-particles above the gap. Since the giant nonlinear conduction was discovered of this material in 2002, we have finally reached a milestone.

4 Pursuing the same dream

The last scientific contact with Tasaki sensei was an international workshop on dynamic cross-effect in softly condensed matter, Tokyo, 4-6 November 2009.[12] This workshop was mainly organized by Professor Yuka Tabé, one of the colleagues in our department. Tasaki sensei and myself were invited there, and discussed the non-equilibrium state of the organic conductor. Although the main participants were soft-matter researchers, unfamiliar with our fields, we received many questions and comments, indicating that Tasaki sensei’s physics included broad interests from physicists. At this workshop, he presented a preliminary work on free energy in a steady state, where his last message was “Yes, we will be almost able to formulate a free energy in current flowing states.” My note for this is shown in Fig. 4.

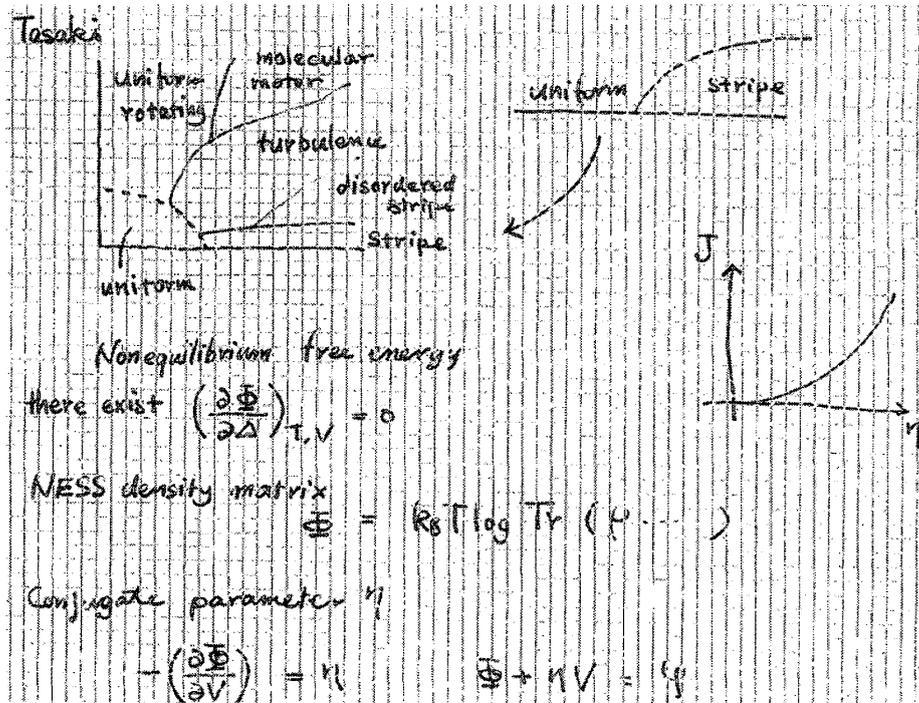


Figure 4: My memo for Tasaki sensei's last work at the international workshop in November 2009.

I really want to see his subsequent work, and heartily regret his loss. So the people reading this article will do. In April 2010 I moved from Waseda to Nagoya University, and extended my research fields. It was just two months since then, when I heard of his death. I often told many colleagues that I had two wings on my shoulders; the one was an excellent x-ray crystallographer, and the other was Tasaki sensei. With the help from the two wings, I could fly higher and higher in the sky of physics. But now, I have lost one of these, being no longer unable to fly...

Although I know that sorrows produce nothing, I still need some time to go further. A small step here in Nagoya is that the current-induced Mott transition in a transition-metal oxide, which occurs at room temperature, well below a metal-insulator transition temperature of 360 K. After the long paper, Ajisaka kun developed his theory where the gap is allowed to change spacially, and found that bipolarons are stabilized under current flow.[13] I am now pursuing this possibility by measuring the optical spectra of the transition-metal oxide under external currents. No one knows that this is the right direction, but I believe that we will eventually reach a meaningful achievement, unless I do not forget the way how Tasaki sensei pinned down the problems. In this respect, I can never thank Tasaki sensei enough for teaching me what a true physics is, and try to go with his enthusiasm for physics through the rest of my research life.

Acknowledgments

I wish to thank the organizers, especially Ajisaka kun, for giving me an opportunity to write this memoir. I would also like to thank the Department of Applied Physics, Waseda University for the free and liberal atmosphere. In such atmosphere, I was able to learn much from Tasaki sensei about physics, mathematics, education, and life.

I understand that this is rather atypical, but highlight the collaboration with Tasaki sensei by the underlines in the following reference list.

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