Unexpected Experimental Results against Common Beliefs Bring Opportunities for New Discoveries

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Overview

Hideki Yamamoto, Senior Distinguished Researcher at NTT Basic Research Laboratories, and his co-researchers are creating novel materials nonexistent in the natural world and elucidating the property of such materials. They apply NTT's unique and state-of-the-art apparatus and technologies to stack the elements making up a material through control of their reaction at the atomic and molecular levels. We asked Dr. Yamamoto about his current research activities and the mindset he adopts as a researcher.

Keywords: superconductor, magnetic material, thin-film synthesis, molecular beam epitaxy

Synthesis of novel materials and discovery of new properties through original and unique methods

—Dr. Yamamoto, please tell us about your current research activities.

My research theme is the design and creation of novel superconducting and magnetic materials through thin-film synthesis methods and explanation of their properties (**Fig. 1**) [1]. The ultimate goals of this research are to contribute to the (i) development of lossless power transmission/supply and wiring, (ii) further reduction in power consumption in devices, and (iii) shift to green-power generation and power storage.

To be more specific, I am involved in creating novel materials not seen in the natural world by growing thin films with a thickness from 0.1 nm (atomic monolayer) to 1 μm on a base of a single crystal (a crystalized solid with regularly arranged atoms) called a substrate. I am also involved in accounting for various, and sometimes novel, properties emerging in such specimens. We use molecular beam epitaxy (MBE) for growing these thin films. With this method, we use ultrahigh-vacuum (UHV) chambers (having a vacuum of one ten-trillionth that of ambient pressure), in which each constituent element of the designated compound is supplied in the form of atoms or molecules to give rise to reaction on heated substrates leading to the formation of thin films [2].

I believe that we are an extremely advanced research team on a global basis working on the creation of completely new compounds by using MBE. Specifically, we are conducting research on thin-film growth of oxides that contain two or more cations (complex oxides) by using a method called oxide

(1) Chemical reactions under low-reaction temperatures and non-equilibrium conditions

Reactions between ultimately small particles such as atoms, molecules, and ions

(2) Stabilization of metastable phases through epitaxy (mutual interaction with the underlying crystal substrate)

(3) Uniform oxidation in the case of oxides (strong oxidation possible through the use of O_3 (ozone) and O)

Tenuity and large surface area to volume ratio of specimens

(4) Impurity-free synthesizing environment (ultrahigh vacuum)

(5) High-throughput syntheses

(6) Resource-saving (usage of small amount of raw material)

(7) Specimens in the form of single-crystal thin films, highly compatible with future device fabrication

Fig. 1. Advantages of searching for and synthesizing new materials by using MBE—a method for growing high-quality thin films of known materials.

MBE. Our custom-made MBE apparatus *sui generis* (**Fig. 2**) has two key features. First, each atomic/molecular flux of constituent cations can be supplied in a stable manner for a prolonged time by monitoring the flux rates of those elements in real time and feeding the results back to the power supply of the evaporation source. Second, it is capable of strong oxidation in a vacuum by supplying oxygen gas, which typically exists as O_2 (oxygen molecules), in the form of highly reactive atomic O (oxygen) or O_3 (ozone) gas. By taking advantage of these features, we have been able to synthesize crystals not found in the natural world.

Using this apparatus, we have been involved for some time in creating new superconducting materials and elucidating their properties. In superconducting materials, electrical resistance becomes zero under certain conditions. In particular, superconductivity occurs at low temperatures below a critical temperature (superconducting transition temperature: T_c). In this state of zero electrical resistance, electricity flows with no power loss within the superconducting material. The highest T_c under ambient pressure (1 atmosphere) is currently –140ºC, which is about 60ºC lower than the temperature at which dry ice sublimes from a solid (approximately –79ºC). However, there have recently been a series of reports on materials having a T_c of –70ºC or even –25ºC closer to room temperature while they can be stable only under ultrahigh pressures (approximately 2 million atmospheres). These reports have led many researchers to believe that room-temperature superconductors must exist but simply have not yet been discovered and synthesized. A report on room-temperature superconductivity in a carbonaceous sulfur hydride, though under 2.7 million atmospheres, has just been published (October 2020) [3]. It strongly suggests that room-temperature superconductivity is no longer an impossible dream.

We, of course, aim to synthesize and discover such materials, but as a prior step, we have been using the thin-film growth methods that I just mentioned to create new superconducting materials having a variety of properties. Examples include the discovery of diverse phenomena such as induction of superconductivity in materials previously thought to be insulators and strain-induced increase in T_c. More recently, we have taken up the challenge of inducing superconductivity in an artificial structure (artificial superlattice) consisting of alternating layers of the simplest building block of cuprate superconductors and oxides containing no copper (Cu); note that the cuprate superconductor family exhibit the highest T_c under ambient pressure. We are now only one step away from superconductivity in such artificial superlattices.

While known as a ferromagnetic metal and neither a new material nor superconductor, we most recently prepared the world's highest quality $SrRuO_3$ (Sr: strontium, Ru: ruthenium) thin film and observed the emergence of an exotic state called a magnetic Weyl semimetal* [4], the quest of which had so far been

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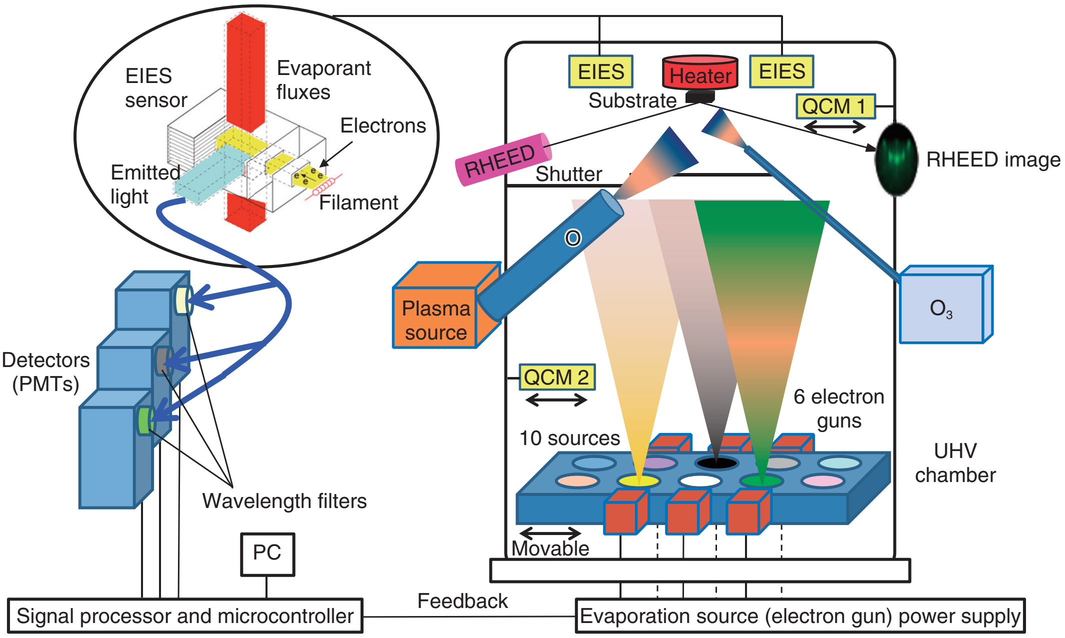
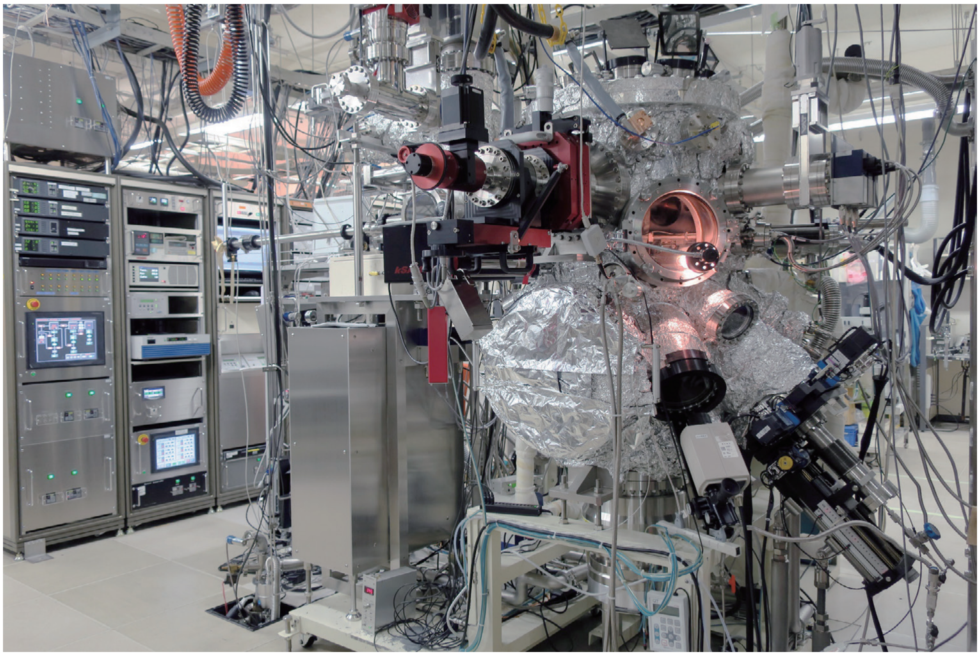
* Magnetic Weyl semimetal: A special state that has only recently been found to occur in some magnetic materials. Weyl quasi-particles that emerge in this state behave as if they have no mass.

(a) Schematic diagram



(b) Photo of the MBE apparatus



EIES: electron impact emission spectroscopy
PC: personal computer
PMT: photomultiplier tube
QCM: quartz crystal microbalance
RHEED: reflection high-energy electron diffraction

Fig. 2. Our MBE apparatus for fabricating complex-oxide thin films.

hampered due to difficulties in preparing high-quality specimens. This accomplishment shows the effectiveness of our high-level thin-film growth technology and measurement techniques for electrical properties under magnetic fields. In future research, we hope to conduct measurements with which the electronic state can be visualized so that the existence of this exotic state can be recognized at a glance.

—Your team is achieving world-class results based on an original method.

The creation of new substances and materials has helped to advance the natural sciences while simultaneously contributing to the development of high-performance and highly functional devices and breakthroughs in device design. This is clearly shown by the discovery of superconductivity in ceramic materials (1987 Nobel Prize in Physics), discovery of fullerenes (molecules consisting of 60 carbon atoms in a soccer ball structure) (1996 Nobel Prize in Chemistry), fabrication of graphene (ultimately thin graphite having a thickness of a single layer of atoms) (2010 Nobel Prize in Physics), and invention of blue light-emitting diodes using a nitride semiconductor (2014 Nobel Prize in Physics). There have also been a variety of proposals and initiatives, in which elements and compounds not previously used will be exploited, for overcoming the limits of miniaturization (the limit of Moore's law) in Si (silicon) integrated circuits, the basis for modern electronics.

Against this background, we have been pursuing the creation of new materials using oxide MBE and successfully created not only superconducting materials but also new magnetic materials. A magnet is a substance whose magnetic property (ferromagnetism) weakens as the temperature increases until eventually disappearing at a certain temperature (Curie temperature). Accordingly, magnetic materials having higher Curie temperature allow for a magnet that can be operated at higher temperatures. With this in mind, we conducted a materials search, which has evolved into the synthesis and discovery of the new ferromagnetic material Sr_3OsO_6 (Os: osmium) [5]. This novel material exhibits the highest Curie temperature (above 780°C) among insulating materials, smashing the record for the first time in 88 years. In addition, Sr_3OsO_6 is free from iron or cobalt unlike most magnetic materials in existence today, which blazes a new trail in the search for magnetic substances. Together with further new materials which, I believe, will be found along the guidelines obtained through the discovery of Sr_3OsO_6, applying these materials to magnetic/spintronic devices that can be operated stably above room temperature will become possible.

—Please tell us how you got started in your world-class research.

In 1987, my former supervisor in this research field moved from Stanford University to NTT Basic Research Laboratories with the aspiration of contributing to both basic and applied research and founded this research unit. About six years later, I entered the laboratories and became involved in this research together with the supervisor and another senior colleague forming a three-person team. At that time, technology for achieving a long-term and stable supply of cation fluxes in a vacuum chamber by providing feedback to the power supply source had not been seen outside of NTT Basic Research Laboratories. I found this to be astonishing, and at the same time, I intuitively felt that this was technology that I should focus on throughout my career. I was fascinated by the possibility of creating novel materials not existing in the world by using this technology.

In 2004, I inherited this research from my former supervisor, and I have since been involved in this work for about 15 years. A variety of experiences have taken place during this time.

We have searched for and synthesized new materials by using the unique thin-film growth techniques that have been cultivated and built up over many years at NTT. However, the manufacturer of the rate controller, which is indispensable for our technologies of high-precision control of the flux rate of each element making up a thin film by electron impact emission spectroscopy (EIES), discontinued the product in 2001. Consequently, thinking that it would be fatal to my research if this technology were to vanish, I consulted with my former supervisor and in the end formulated a specification and ordered a custom-made product from a startup company in Silicon Valley, USA. For about one year starting about three months after that order, I had the good fortune of researching at Stanford University for joint research, which allowed me to visit that company frequently during breaks in my research. Thanks to this state of affairs, I was able to personally participate in the completion of this custom-made product from the operation-verification stage of the first prototype (**Fig. 3**). This technology has further evolved over the 15 years since the development of the prototype, and

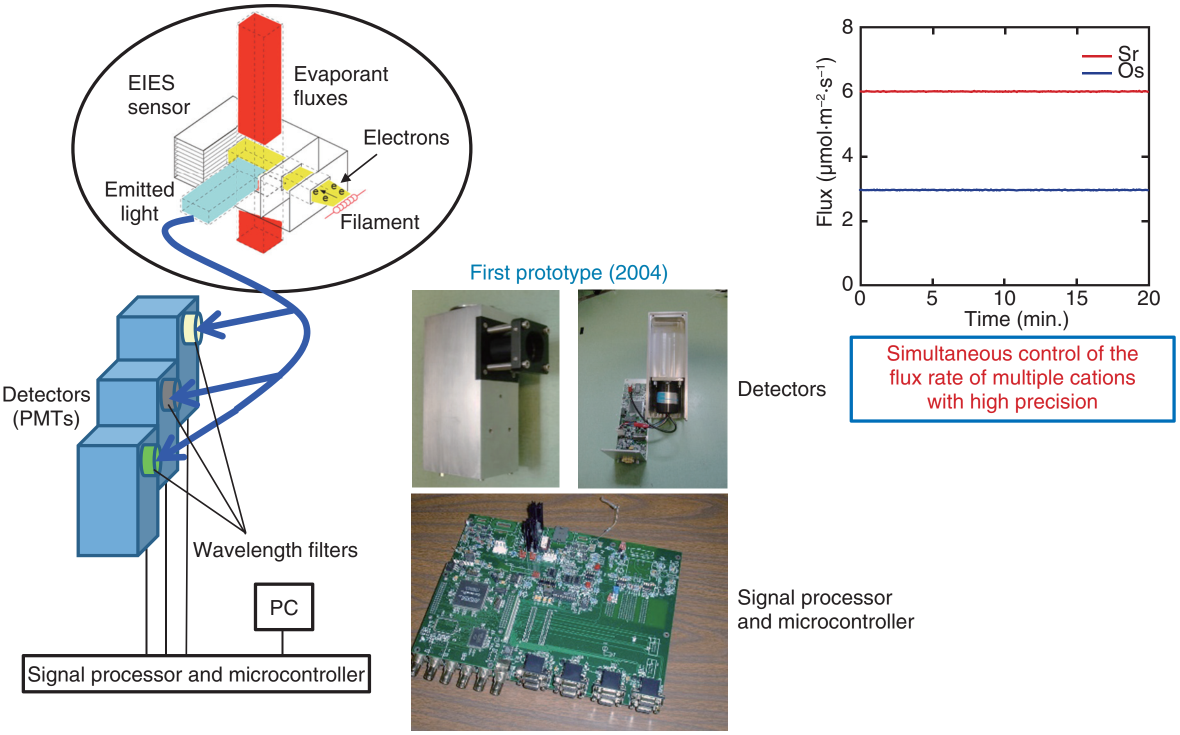


Fig. 3. Flux-rate control equipment using EIES (first prototype of custom-made product).

looking at the way that it has supported our research is deeply moving for me.

When involved in research, there are tough years to contend with, but also exciting years when discoveries, even if small, occur one after another. My first thrilling experience occurred in my third year at NTT. I was able to interpret the experimental results from the research that I had taken up immediately after entering the company and to then publish my first paper on those results. My good fortune then continued with our discovery of the first new superconductor synthesized using the thin-film growth method and other achievements. Our research team was given a boost when my senior colleague received the Young Scientist Presentation Award of the Japan Society of Applied Physics in 1996 and I the same award the following year. On the other hand, I remember well how our supervisor at that time admonished us young researchers excited about our string of successes with the words "perform your experiments simply and honestly." Experimental science is the process of understanding nature by changing experimental conditions in a variety of ways to query nature on its laws and using the answers received. For this reason, the significance of the words "simply and honestly" spoken by him sticks with me to this very day and at every turn.

In subsequent years, more encouragement came my way by receiving the Superconductivity Science and Technology Award (2016) that I had privately longed for and by having the opportunity to publish a number of papers in leading journals. More recently, however, I am frequently called upon to submit scientific manuscripts as the last author (usually, the person in charge of those research projects), so it's more a sense of relief than a feeling of joy when we've gotten a paper published. Within the laboratories, I sometimes hear young researchers talk about their tears of joy over getting a paper published, which I feel a bit envious about. I guess this is a clear sign of my age!

Setting big goals (high mountains to climb) and taking a safe and steady approach

—How have your views of things and role changed after becoming a senior distinguished researcher?

I am grateful for being put into a stable position that allows me to concentrate on my research. I believe I can take appropriate and legitimate risks for making breakthroughs and acquiring high returns by exploiting this position. Taking risks is vital to achieving breakthroughs in basic research, especially in materials research. However, there are many cases in which there is no other choice but to select research themes for which results and papers can be readily produced to secure the next positions of team members and win competitive research funding. The reality is that taking risks is not easy. I therefore want to make good use of my very favorable position and environment as an NTT senior distinguished researcher to pursue research that takes appropriate and legitimate risks.

Among the 11 core technologies for making the NTT vision of the Innovative Optical and Wireless Network (IOWN) a reality, 6 are related in some form to materials research, so in this sense too, I feel the weight of responsibility. Although my theme of design and thin-film synthesis of novel superconductors and magnetic materials with elucidation of the underlying physics is not necessarily a mainstream one in IOWN, there are many examples in history of major breakthroughs coming out of non-mainstream research themes, and I have expectations in this regard.

I also feel that forming research teams that are diverse not only in gender, nationality, and language but also in expertise is important for achieving breakthroughs. It is easy to form and manage a research team whose members are in the same field of expertise, but I think it's difficult for such a team to make major breakthroughs. I think it's great if a young researcher who joined a research team but with a different field of expertise occasionally thinks, "Why don't my seniors know about this?" From a short-term perspective, many points of difference can make things all the more difficult, but from a long-term perspective, the importance of such diversity will be felt.

—Is there anything that you have kept in mind when searching for problems or themes to work on?

Since entering NTT Basic Research Laboratories after receiving my doctorate, setting big goals (high mountains to climb) and taking a safe and steady approach is something that I have always kept in mind. For example, the search for room-temperature superconductors included in my research theme is a high mountain to climb and an appropriate theme for a researcher in my present position. The search for artificial photosynthetic material on par with plants is likewise a high mountain to climb. However, when you're young and have a dream, you might take on a significant challenge from the beginning, and I was the same way. This can lead to a situation of trying to push a massive rock with all one's might and failing to move it even 1 mm. To avoid taking such an all-or-nothing approach, it is advisable to achieve small milestones and write papers on those results as they happen. I learned this approach from my seniors at NTT Basic Research Laboratories.

Basic research does not go well if the theme is one that the researcher does not truly enjoy. It is not always a short run to the top of the mountain, and it is not unusual to have to take detours or pull back as needed. In such circumstances, it is important to adopt a mindset in which you never quit looking up at the top of the mountain. Good basic research can have a major impact on creating or changing a certain concept. In this sense, an example of good basic research that I'm particularly proud of was our research of novel superconducting materials that we synthesized and discovered over about ten years from 2003. This research presented counterexamples to previously established superconductivity emergence conditions in cuprate superconductors. I felt disheartened when our paper was initially not accepted by academic journals, but at least the research was a thrilling experience. Although these materials were established as novel superconductors, a dispute as to whether our interpretation of the superconductivity-emergence mechanism is truly a counterexample to the established theory is continuing. This is also a common occurrence in basic research, so I maintain a positive attitude.

Imagining one's ideal research life given ample funding

—What would you like to say to junior researchers?

Materials informatics has recently appeared as the fourth paradigm of science after experimentation, theory, and computational science. At the same time, the accuracy of predicting the electronic structure within materials through theoretical computations has significantly improved. As a result, it has become possible to conduct materials search in a considerably more efficient manner than before and optimize thin-film growth conditions. I would like junior researchers to use such efficient methods and avoid the detours that I took due to my own lack of ability. However, the reality is that there are still many things you won't understand unless you actually conduct experiments in materials science, so I would like junior researchers to keep in mind the following advice.

Before doing anything else, construct a high-reliability experimental system that can reproduce experimental results. Once you complete this to a reasonable extent, try conducting some experiments. It's important to pursue experiments with good efficiency, but it's also important to try detours without emphasizing only *short-distance runs*. While the reproduction of results is a prerequisite in science, experimental results that differ from what was expected or from commonly accepted theory present a great opportunity. I believe that there's a strong possibility of encountering an unknown or unexpected phenomenon through those experimental results. Moreover, experimental results thought to be a failure in the sense of achieving a certain goal, cannot alone be judged a failure. For example, while it would certainly be disappointing to fail in preparing a thin film with the target level of quality, it is not uncommon for some hints to be hidden in those experimental results.

I sometimes ask young researchers, "What kind of research life would you like to lead and what kind of research would you like to pursue if your pay was enough to lead a worry-free life and you could receive ample funding for research?" The reply to the question should be, "the research that I have a great desire to pursue." In the long run, it would be better to pursue a research theme determined in this manner.

As you know, originality is more important than anything else in basic research. On the other hand, a former supervisor of mine once told me that research on current technology is not something to detest as long as it concerns truly important technology. Avoiding researching current technology and falling to a technological level at which you cannot even reproduce fascinating results that some other research groups have provided is worse than researching it. I believe that you have sound science if multiple top-level research teams and institutions can reproduce good basic research results.

—Dr. Yamamoto, can you tell us about your future research topics?

Until several years ago, the elements that we used to grow thin films of complex oxides included transition metals on the relatively upper rows of the periodic table such as Cu. Recently, however, we have expanded our target to transition metals such as Ru, Pd (palladium), and Os that belong to lower rows than that of Cu on the periodic table, which, for example, has led to the discovery of a material having magnetic properties up to the highest temperatures among insulators. Our next theme is to what extent we can systematically and strategically (but not haphazardly) expand our use of these transition metals. In addition, the discovery of materials that can exist stably only under ultrahigh pressures of 2 million atmospheres but make a superconducting transition at temperatures closer to room temperature has been reported for hydrides, not oxides. Up to now, our target for growing thin films has been essentially limited to complex oxides, so how we should expand our target beyond oxides is also in my things-to-do list.

Tackling these issues is going to take some time—it is not something that can be completed within one's own generation. Training up-and-coming researchers who will become the principal investigators of next-generation research while continuing to produce research results is of prime importance. I feel the need for passing on knowledge as well as technical skills. This is necessary not only for our researchers but also for equipment and component manufacturers that support our research, so I think it would be worthwhile to develop some means of passing on technology on both sides.

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■ Interviewee profile

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He received a B.S., M.S., and Ph.D. in chemistry from the University of Tokyo in 1990, 1992, and 1995. He joined NTT in 1995, and his principal research fields are thin-film growth, surface science, and condensed-matter physics. He was a visiting scholar at the Geballe Laboratory for Advanced Materials, Stanford University, USA (2004–2005). He received the 2nd Young Scientist Presentation Award (1997) from the Japan Society of Applied Physics (JSAP) and the 20th Superconductivity Science and Technology Award (2016) from the Forum of Superconductivity Science and Technology, the Society of Non-traditional Technology. He is a member of JSAP, the Physical Society of Japan, the Japan Society of Vacuum and Surface Science, the American Physical Society, and the Materials Research Society.