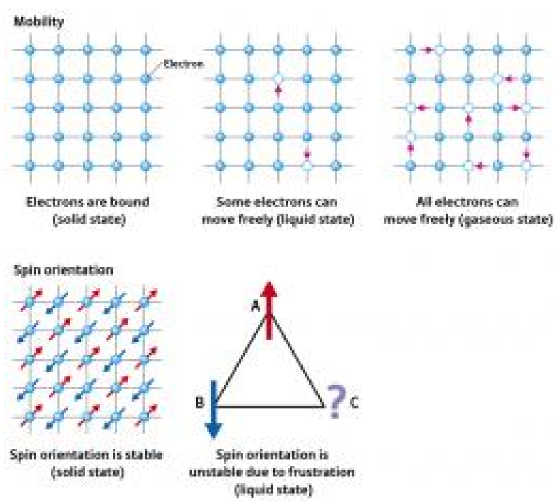
Using complex electron systems to create green materials

January 31 2011, By Hidenori Takagi



State change in complex electron systems. Complex electron systems can change into various states depending on the entanglement of electrons (upper) or spin orientation (lower)

At the L'Aquila Summit held in July 2009, G8 nations reaffirmed the target of at least a 50% reduction in greenhouse gas emissions worldwide by 2050. They also set a target for developed countries to reduce greenhouse gas emissions by 80% or more. To achieve these targets, it is imperative to not only improve existing technologies, but also develop innovative technologies based on new principles.

Accordingly, the RIKEN Advanced Science Institute (Japan) established the Green-forefront Materials Department in April 2010, aiming to create 'green-forefront materials', that is, new materials for helping to solve environmental and energy problems. In the same year, Hidenori Takagi set up and led the Complex Electrons and Functional Materials Research Group in the Green-forefront Materials Department. This group is striving to create high-temperature superconductors and thermoelectric materials using complex electron systems.

This year will mark the 100th anniversary of the discovery of the superconductor by the Dutch physicist Heike Kamerlingh Onnes. Superconductors are materials in which electrons can flow with no electrical resistance, and the first such materials displayed this behavior when cooled to near absolute zero. This year will also mark the 25th anniversary of the discovery of the first 'high-temperature' cuprate superconductor—a material that makes the transition to the zero-resistance state at a temperature of 30 K. In that year, 1986, Takagi successfully conducted a follow-up experiment to verify the discovery of the high-temperature cuprate superconductor. A worldwide search for high-temperature superconductors then ensued with the hope of discovering new materials that could significantly contribute to solving environmental and energy problems as a medium for lossless electric power transmission and storage. Today, the highest transition temperature achieved to date for cuprate superconductors is 160 K, or about −113 °C, under high pressure.

Cuprates are basically insulators, so it is one of the most intriguing questions in science how such materials can become superconductors. According to Takagi, "Metals contain unbound electrons that can move freely through the material, so when a voltage is applied to these electrons in this 'gaseous' state, an electrical current flows." Free electrons are also present in some transition-metal elements such as cuprates (metallic oxides including metals from group III to group II in the periodic table). "Their electron orbits, however, are so narrow that they cannot move to adjacent atom because they are electrically repelled by the existing electrons of the atom. This state can be compared to that of the solid state in which electrons are stuck tightly in place because they repel each other. The material in this state acts as an insulator. Systems composed of such strongly interacting electron populations are known as strongly correlated electron systems or complex electron systems, and they can change their electrical states or electron phases dramatically in response to small changes in conditions."

For example, as electron vacancies, called holes, are created in such systems, the remaining electrons slowly begin to move, changing the electron state from tightly bound solid state to more fluid mobile state (Fig. 1). "In strongly correlated electron systems, the solid electron state first changes into a sticky liquid and then a gaseous state in which electrons can move freely. These systems change state in response to changes in conditions. That is where the fun is."

High-temperature superconductivity occurs when complex electron systems change state. Electrons in a superconductor are known to form pairs of electrons called Cooper pairs. The mechanism by which electrons form Cooper pairs in cuprates when they usually repel each other under, however, remains a mystery. Takagi and his group have been conducting world-leading research into achieving direct observations of the electron state in high-temperature cuprate superconductors by scanning tunneling microscopy (STM). "We found that most electrons in high-temperature cuprate superconductors are stuck tightly in place except for a small number of electrons that are left free from atomic bonds. This state is considered to be what creates Cooper pairs and causes superconductivity.

Many unknown electron states lie hidden in complex electron systems. "We aim to discover new electron states and to use them to create new materials."

The states of complex electron systems can also be changed by taking advantage of the property of electrons known as 'spin'. Spin describes the angular momentum of an electron, similar in nature to the rotation of Earth, and can be either 'up' or 'down'. In the same was as two bar magnets arranged in the same direction repel each other yet when arranged in opposite directions are pulled together, a square lattice of atoms is most stable when the up and down spins of adjacent are arranged alternately (Fig. 1). "If the directions of electron spins are arranged in an orderly manner, spins are considered to form a spin solid. How are the spins aligned in a triangular atomic lattice? There are three electrons located at the vertices of a triangle, but only two spins can form a pair, leaving one unpaired spin. In such a lattice, the orientations of electron spins are unstable. This state, called 'spin frustration', is considered to be responsible for s spin liquid state."

Electron systems tend to become more stable at lower temperature. In 1973, however, it was theoretically predicted that a lattice with extremely strong spin frustration could adopt a special state called a 'quantum spin liquid', in which the directions of electron spins do not become stable even when the temperature is lower to absolute zero. "This state, of course, is very hard to produce and has long been considered merely a theoretical dream."

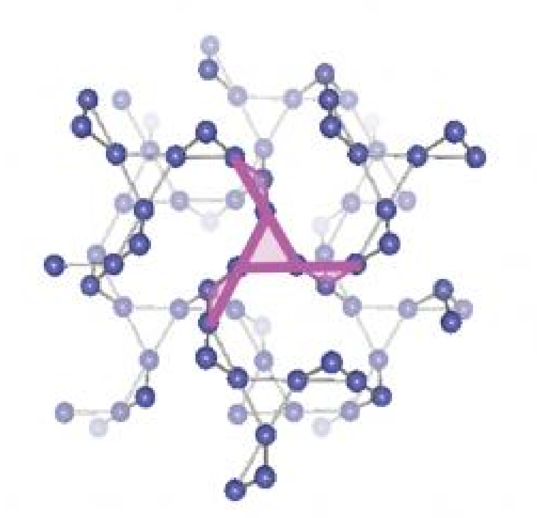


Figure 2: $Na_4Ir_3O_8$, a transition-metal oxide that can turn into a quantum spin liquid. The transition-metal oxide $Na_4Ir_3O_8$ has a triangular lattice that forms spirals. It exhibits strong spin frustration and becomes a quantum spin liquid at absolute zero.

Yet Takagi and his group were eventually successfully in creating a quantum spin liquid using the compound $Na_4Ir_3O_8$, a transition metal oxide (Fig. 2) consisting of a triangular lattice of atoms in a spiral configuration. Takagi named this structure the 'hyper-kagome lattice'. Another research group also successfully created a quantum spin liquid using a different structure at around the same time, and the field now attracts a great deal of attention.

The possible uses of complex electron systems are actually more practical than might be expected. "One of the best examples is water ice that freezes at 10 °C," says Takagi. "Wine and sushi, for example can be overcooled if placed in normal ice at 0 °C. Ice that freezes at 10 °C would be very convenient in these types of situations. We have in fact created a coolant in which the electron state changes from solid to liquid at 10 °C."

Takagi and his co-workers have also taken advantage of spin frustration to successfully develop a new compound known as Mn3XN with a thermal expansion coefficient of zero (Fig. 3). Manganese nitride, the base material for the new compound, has a triangular lattice of manganese atoms. The compound adopts a liquid state at high temperature because of its electron state, and becomes a solid when the temperature is lowered to near room temperature. The electrons try to stay at the triangular lattice points, but the directions of electron spins remain unstable because of the strong spin frustration in this system. The surrounding lattice, however, expands when cooled to relax the spin frustration, resulting in a material that expands when cooled and contracts when heated—the opposite behavior to that of most natural compounds. For example, an iron bar increases in length by 0.0012% for every 1 °C increase in temperature. Although small, such thermal expansion can sometimes causes problems in processing equipment and high-precision measuring devices, particularly in semiconductor circuits where nanometer-accuracy is required.

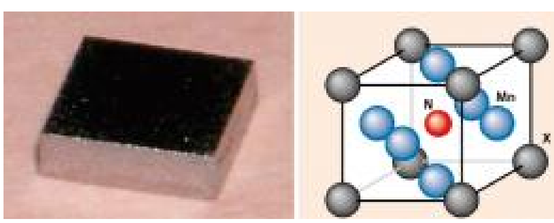


Figure 3: Mn3XN, a manganese nitride with negative thermal expansion coefficient. The manganese nitride Mn3XN has an antiperovskite structure. The symbol X represents copper or zinc. The component X can be replaced with germanium or tin atom, resulting in a material with a zero thermal expansion coefficient.

“Manganese nitride itself contracts discontinuously at a certain temperature. By introducing germanium or tin, we can create a substance that contracts gradually over a temperature range of about 100 °C. In 2008, we successfully created a new material with a zero thermal expansion coefficient, meaning it does not change volume, over a temperature range of about 70 °C including room temperature."

Conventional substances with a zero thermal expansion coefficient are compound materials consisting of compounds with different expansion coefficients, which leads to deformation or cracking and low strength. Such materials are also usually expensive because they contain rare elements. "Manganese nitride is resistant to deformation and cracking, and is also less expensive than conventional substances because it is a single compound."

In 2010, Takagi and his colleagues started the Complex Electrons and Functional Materials Research Group at the RIKEN Advanced Science Institute. "We aim to create 'green-forefront materials' that help to solve environmental and energy problems by taking advantage of complex electron systems. A typical example of our work is the development of high-efficiency thermal conversion materials."

Only 30% of the thermal energy produced by the combustion of gasoline in an automobile engine is used to power the vehicle; the other 70% is lost as waste heat. A large proportion of the thermal energy produced by burning fossil fuels in thermal power stations and many factories is also lost as waste heat. The key to reducing fossil fuel usage and carbon dioxide emissions is how effectively that waste heat can be harnessed. There are high hopes for thermal conversion materials, or thermoelectrics, which convert heat energy into electrical energy and vice versa.

"The currently used Bi2Te3-based thermoelectrics cannot be used at high temperatures and are not very efficient at converting heat to electricity. These include the Peltier elements used in some mobile refrigerators, which use electricity to cool beverages, for example, by the reverse thermoelectric effect, but which do not have sufficient conversion efficiency to make ice. We aim to develop efficient thermoelectric materials that could be used to freeze water.”

Spin frustration could be utilized to increase the thermal conversion efficiency of such materials. “The principle of heat-to-electricity conversion is based on temperature difference. A temperature difference causes electrons or holes to move and thus produce a voltage, generating electrical power. The higher the disorder in the electron state, the higher the conversion efficiency, and the more electricity is generated. For example, entangled electrons with strong spin frustration are unstable and move in a more random manner, increasing the amount of disorder, or entropy.” Takagi and his team are working to create new high-efficiency thermoelectrics based on a new principle such as spin frustration.

In order to efficiently use the waste heat generated by vehicles and factories, the thermoelectric materials must be able to withstand temperatures of more than several hundred degrees celsius. “We should be able to develop heat-resistant, high-efficiency thermoelectrics because the transition metal oxides we are dealing with are strong and heat-resistant.”

In 2008, Hideo Hosono and his research group at the Tokyo Institute of Technology in Japan discovered an iron-based superconductor that attracted worldwide attention. “This superconductor has a transition temperature of 55 K, which is currently the highest transition temperature for non-cuprates. It was previously considered impossible to produce magnetic superconducting materials such as those containing iron, cobalt or nickel, so these iron-based superconductors probably have a different superconducting mechanism to that in cuprates,” says Takagi.

In April 2010, Tetsuro Hanaguri, a senior research scientist in RIKEN's Magnetic Materials Laboratory, successfully observed the structure of a Cooper pair of electrons in an iron-based superconductor by STM. Metallic superconductors are known to form a type of Cooper pair called an s wave, whereas cuprate superconductors form a Cooper pair called a d wave. Hanaguri demonstrated for the first time that iron-based superconductors formed a new type of Cooper pair called an s± wave. Spin fluctuation is considered to be the mechanism responsible for creating Cooper pairs in iron-based superconductors.

Recent advances in research on superconductivity have raised the expectations for creating room-temperature superconductors, which would be the ultimate green forefront material. "The highest-temperature superconductor produced so far was discovered in 1994, a record that has remained unbroken for 16 years. This is the longest period since 1911, when superconductivity was first discovered, that there has been no increase the highest recorded transition temperature. We are adrift in research on higher-temperature superconductors. To challenge the status quo, I would like to break the long-standing record of 160 K by discovering a new high-temperature superconductor based on a new principle, which would again trigger a worldwide race for higher-temperature superconductors."

Provided by RIKEN

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