Tuning into the LCDs of tomorrow: Exploring the novel IGZO-11 semiconductor

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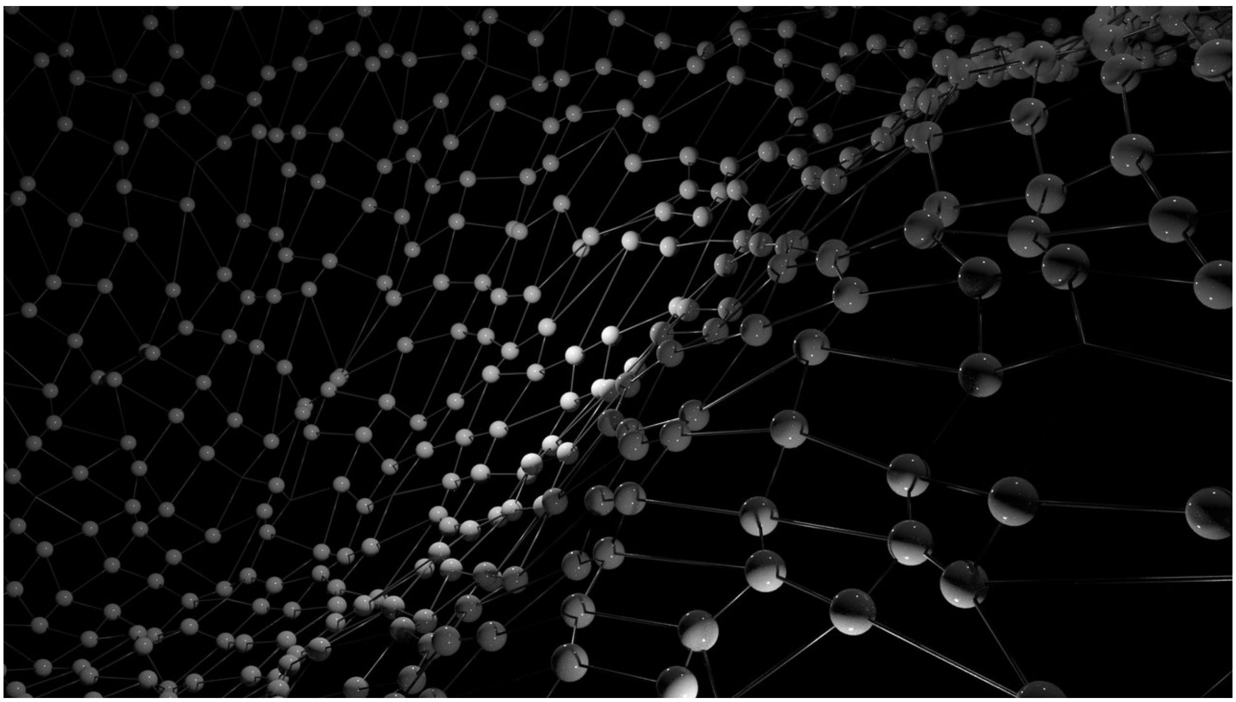

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In 1985, Noboru Kimizuka of the National Institute for Research in Inorganic Materials, Japan had pioneered the idea of polycrystalline indium-gallium-zinc oxide (IGZO) ceramics, with the general chemical formula ($InGaO_3$)m(ZnO)n (m, n = natural number; hereafter referred to as IGZO-mn). Little would he have thought that its curious electrical properties would bring the electronics industry to license thin-film transistors (TFTs) made from these metal oxides for various devices, including touchable displays. However, this did not come easy. Even today, many of the characteristics of pure IGZO crystals remain unknown owing to their difficult extraction procedure. Then what makes them tantalizing?

When you shine light on metals, the free conducting electrons resonate or vibrate with external light (electromagnetic waves). Thus, the light wave is shielded, and as a result, light is not transmitted but is reflected. This is why metals are not generally transparent despite being good reflectors and conductors. In contrast, semiconductors with a large band gap, such as IGZO, can absorb and transmit light even in the visible light range. In general, the large band gap implies that these types of materials are insulators. Injecting carriers, using oxygen defects, into a semiconductor material with a large band gap can yield a material that is both transparent and conductive.

Thus, being both transparent and conductive makes these semiconductors suitable for use in optoelectronic devices, much like the one you're reading this on! Furthermore, IGZO-based transistors have added advantages such as high electron mobility, good uniformity over a large area, and low processing temperature, which make it possible to achieve unparalleled energy-efficient high resolution. Within this IGZO-1n family, polycrystalline IGZO-11 (i.e. $InGaZnO_4$) exhibits the highest conductivity and the largest optical band gap. In addition, von Neumann-type computers, or simply digital computers, require "on-off state" electrical circuits as the basic building blocks, with the ideal "off" state corresponding to a "zero" current. The IGZO-11 excels on this front too, as the off-state current value for it is extremely small, which implies that the energy loss can be minimized.

However, sufficiently big single crystals of IGZO-11 that could be used to measure their physical properties have not yet been obtained.

Therefore, its precise intrinsic properties are unexplored. Motivated by this and the fact that a multicomponent oxide with a layered structure could exhibit anisotropic conduction, a team of researchers, mainly from Tokyo University of Science, led by Prof Miyakawa, has developed a novel technique to grow single crystals of the type.

The primary challenge in the synthesis of the multicomponent layered structure is the recurrent defect formation during crystal growth. Furthermore, the physical properties of the material were unknown, which meant that the route for isolating the crystal had to be meticulously chalked out. Faced with the fact that the IGZO-11 might also be an incongruent material under atmospheric pressure (i.e., the crystalline solid phase is decomposed in the melting process into a second crystal phase, different from the original crystal, and a liquid phase), the research team opted for optical floating zone (OFZ) to build the crystal. By increasing the gas pressure, the team succeeded in suppressing the evaporation and vaporization, and growing a good single crystal from the liquid phase.

Thus, OFZ enabled the growth of high-quality oxide crystals without the need for a crucible or a container, which gives better control over the temperature and pressure that the liquid material is subjected to. Additionally, the use of Zn-rich feed-rod in the synthesis allowed the researchers to control the level of ZnO that would have otherwise evaporated, rendering the synthesis futile.

Upon succeeding with the synthesis of the crystal, the researchers studied its physical properties. They observed that the nascent crystal appeared bluish in color. On annealing or heating and then slowly cooling in free atmosphere and additional oxygen, the crystal became transparent. Free carriers produced by oxygen vacancies in crystals absorb red light and emit blue light; thus, the researchers associated the color change with this oxygen filling the vacancies when the crystal underwent annealing.

To complete the tale, the researchers then measured the crystal's electrical conductivity, mobility, and carrier density, and their temperature dependences. They noted that post-annealing all electrical properties showed a decrease. The carrier density and conductivity could be controlled within the range of 10^{17} to 10^{20} cm^{-3} and 2000-1 S cm^{-1} at room temperature by post-annealing. They also reported an increase in the mobility upon increase in carrier density, which was previously noted in transport studies for some IGZO-1n thin films. This suggests that the unusual behavior is an intrinsic characteristic of the IGZO-1n family.

Interestingly, the team noted that the conductivity along the c-axis (axis perpendicular to each plane in the layered structure) is >40 times lower than that in the ab-plane (plane of the layer) in the single crystals, and that the anisotropy increases with decreasing carrier density. As Prof Miyakawa explains, "Indium-indium distance along the c-axis is much longer than that along the ab-plane. Therefore, the overlap of the wave function is smaller in c-axis direction." Because the degree of overlap of the wave-functions of electronic orbitals governs how easily electrons can move, the researchers assert that this could be the origin of the anisotropic conductivity for IGZO-11 crystals.

Previously, the IGZO family has been used in liquid crystal displays, including in smartphones and tablets and, in fact, recently also in large OLED televisions. The electrical conductivity and transparency of this novel material make IGZO stand out. While fabricating transistors out of the IGZO-11 that can be directly applied in LEDs remains a work in progress, this fascinating research marks the start of many more discoveries.

So, do you see why IGZO-11 is important or are you seeing through it?

More information: Yusuke Tanaka et al, Single crystal growth of bulk InGaZnO4 and analysis of its intrinsic transport properties, *CrystEngComm* (2019). DOI: 10.1039/C9CE00007K

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