Uncovering oxygen's role in enhancing red LEDs

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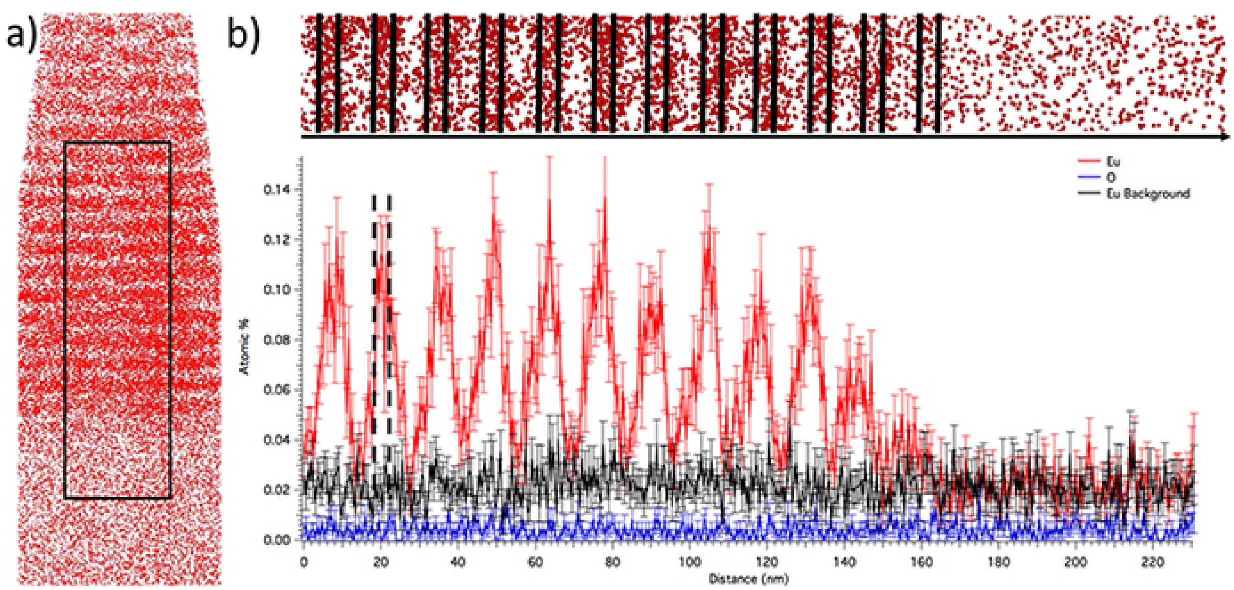

A) shows the europium (Eu) distribution of the delta structure (DS) samples with alternating 10-nanometer gallium nitride (GaN) layers and 4-nm GaN:Eu layers. A zoomed in view (b) of the DS sample structure aligns with a plot of the atomic percentage of Eu and oxygen as a function of space. The background signal of Eu is also indicated for reference. Credit: B. Mitchell, D. Timmerman, J. Poplawsky, W. Zhu, D. Lee, R. Wakamatsu, J. Takatsu, M. Matsuda, W. Guo, K. Lorenz, E. Alves, A. Koizumi, V. Dierolf & Y. Fujiwara

Oxygen is indispensable to animal and plant life, but its presence in the wrong places can feed a fire and cause iron to rust.

In the fabrication of solid state lighting devices, scientists are learning, oxygen also plays a two-edged role. While oxygen can impede the effectiveness of gallium nitride (GaN), an enabling material for LEDs, small amounts of oxygen in some cases are needed to enhance the devices' optical properties. GaN doped with europium (Eu), which could provide the red color in LEDs and other displays, is one such case.

Last week, an international group of researchers shed light on this seeming contradiction and reported that the quantity and location of oxygen in GaN can be fine-tuned to improve the optical performance of Eu-doped GaN devices. The group includes researchers from Lehigh, Osaka University in Japan, the Instituto Superior Técnico in Portugal, the University of Mount Union in Ohio, and Oak Ridge National Laboratory in Tennessee.

Writing in *Scientific Reports*, a Nature publication, the group said that small quantities of oxygen promote the uniform incorporation of Eu into the crystal lattices of GaN. The group also demonstrated a method of incorporating Eu uniformly that utilizes only the oxygen levels that are inevitably present in the GaN anyway. Eu, a rare earth (RE) element, is added to GaN as a "dopant" to provide highly efficient red color emission, which is still a challenge for GaN-based optoelectronic devices.

The devices' ability to emit light is dependent on the relative homogeneity of Eu incorporation, said Volkmar Dierolf, professor and chair of Lehigh's physics department.

"Some details, such as why the oxygen is needed for Eu incorporation, are still unclear," said Dierolf, "but we have determined that the amount required is roughly 2 percent of the amount of Eu ions. For every 100 Eu ions, you need two oxygen atoms to facilitate the incorporation of Eu to GaN.

"If the oxygen is not there, the Eu clusters up and does not incorporate. When the oxygen is present at about 2 percent, oxygen passivation takes place, allowing the Eu to incorporate into the GaN without clustering."

The article is titled "Utilization of native oxygen in Eu(RE)-doped GaN for enabling device compatibility in optoelectronic applications." The lead author, Brandon Mitchell, received his Ph.D. from Lehigh in 2014 and is now an assistant professor of physics and astronomy at the University of Mount Union and a visiting professor at Osaka University.

Coauthors of the article include Dierolf; Yasufumi Fujiwara, a professor of materials science at Osaka University; and Jonathan D. Poplawsky, a research associate at Oak Ridge National Laboratory who received his Ph.D. from Lehigh in 2012.

A comprehensive study

Gallium nitride, a hard and durable semiconductor, is valued in solid state lighting because it emits light in the visible spectrum and because its wide band gap makes GaN electronic devices more powerful and energy-efficient than devices made of silicon and other semiconductors.

The adverse effect of oxygen on GaN's properties has been much discussed in the scientific literature, the researchers wrote in *Scientific Reports*, but oxygen's influence on, and interaction with, RE dopants in GaN is less well understood.

"The presence of oxygen in GaN," the group wrote in their article, which was published online Jan. 4, "...is normally discussed with a purely negative connotation, where possible positive aspects of its influence are not considered.

"For the continued optimization of this material, the positive and negative roles of critical defects, such as oxygen, need to be explored."

The group used several imaging techniques, including Rutherford Backscattering, Atomic Probe Tomography and Combined Excitation Emission Spectroscopy, to obtain an atomic-level view of the diffusion and local concentrations of oxygen and Eu in the GaN crystal lattice.

Its investigation, the group wrote, represented the "first comprehensive study of the critical role that oxygen has on Eu in GaN." The group chose to experiment with Eu-doped GaN (GaN:Eu), said Dierolf, because europium emits bright light in the red portion of the electromagnetic spectrum, a promising quality given the difficulty scientists have encountered in realizing red LED light.

The group said its results "strongly indicate that for single layers of GaN:Eu, significant concentrations of oxygen are required to ensure uniform Eu incorporation and favorable optical properties.

"However, for the high performance and reliability of GaN-based devices, the minimization of oxygen is essential. It is clear that these two requirements are not mutually compatible."

Preliminary LED devices containing a single 300-nanometer active GaN:Eu layer have been demonstrated in recent years, the group reported, but have not yet achieved commercial viability, in part because of the incompatibility of oxygen with GaN.

To overcome that hurdle, said Dierolf, the researchers decided that instead of growing one thick, homogeneous layer of GaN:Eu they would grow several thinner layers of alternating doped and undoped regions. This approach, they found, utilizes the relatively small amount of oxygen that is naturally present in GaN grown with organo-metallic vapor phase epitaxy (OMVPE), the common method of preparing GaN.

"Instead of growing a thick layer of Eu-doped GaN," said Dierolf, "we grew a layer that alternated doped and undoped regions. Through the diffusion of the europium ion, oxygen from the undoped regions was utilized to incorporate the Eu into the GaN. The europium then diffused into the undoped regions."

To determine the optimal amount of oxygen needed to circumvent the oxygen-GaN incompatibility, the researchers also conducted experiments on GaN grown with an Eu "precursor" containing oxygen and on GaN intentionally doped with argon-diluted oxygen.

They found that the OMVPE- grown GaN contained significantly less oxygen than the other samples.

"The concentration of this oxygen [in the OMVPE- grown GaN] is over two orders of magnitude lower than those [concentrations] found in the samples grown with the oxygen-containing Eu...precursor," the group wrote, "rendering the material compatible with current GaN-based devices.

"We have demonstrated that the oxygen concentration in GaN:Eu materials can be reduced to a device-compatible level. Periodic optimization of the concentration ratio between the normally occurring oxygen found in GaN and the Eu ions resulted in uniform Eu incorporation, without sacrificing emission intensity.

"These results appear to coincide with observations in other RE-doped GaN materials. Adoption of the methods discussed in this article could have a profound influence on the future optimization of these systems as well as GaN:Eu."

The group plans next to grow GaN quantum well structures and determine if they enable Eu to incorporate even more favorably and effectively into GaN. Toward that end, Dierolf and Nelson Tansu, professor of electrical and computer engineering and director of Lehigh's Center for Photonics and Nanoelectronics, have been awarded a Collaborative Research Opportunity (CORE) grant from Lehigh.

More information: B. Mitchell et al. Utilization of native oxygen in Eu(RE)-doped GaN for enabling device compatibility in optoelectronic applications, *Scientific Reports* (2016). DOI: 10.1038/srep18808

Provided by Lehigh University

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