Researchers use quantum computer to identify molecular candidate for development of more efficient solar cells

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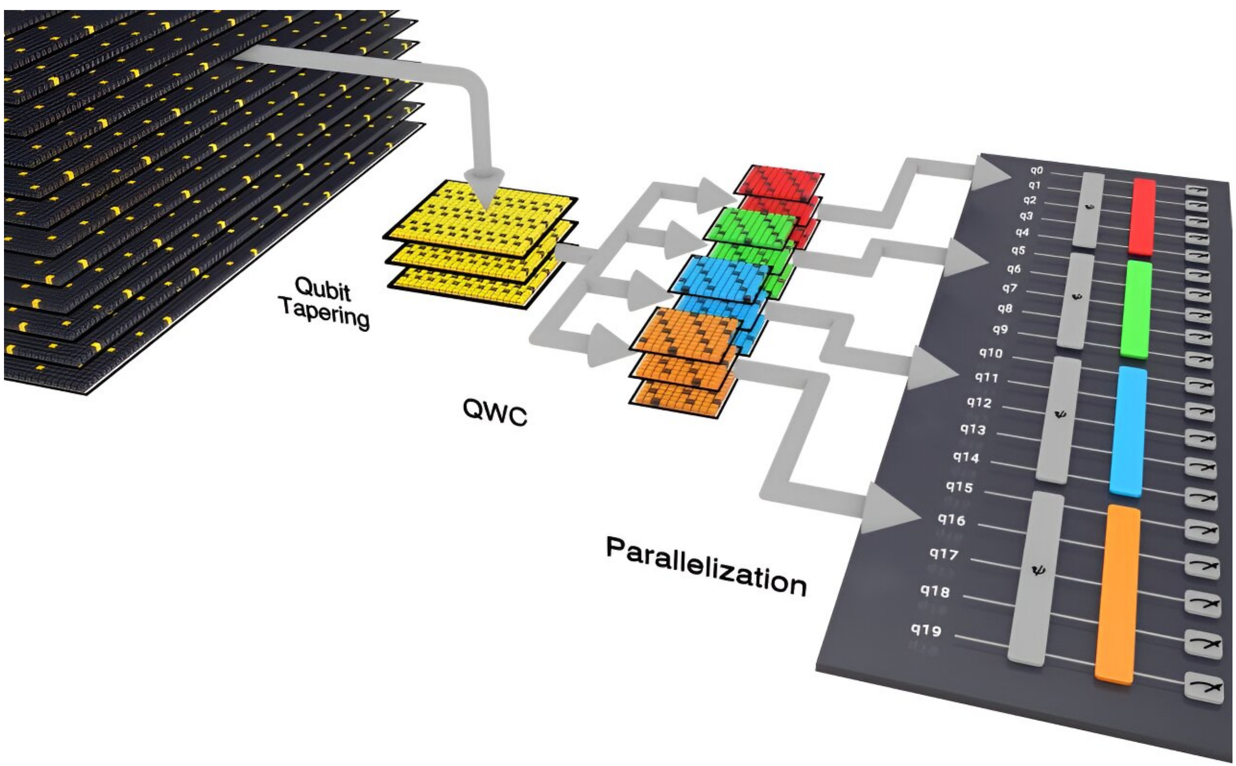

ORNL team members applied three independent strategies to decrease their project's computational workload, which reduced their time to solution from months to a few weeks. First, in a technique called qubit tapering, they decreased the number of qubits required to express the problem, reducing the size of the problem itself. Second, they took fewer measurements to solve the problem by measuring groups of terms once rather than measuring each individual term from every group (a process called qubit-wise commutativity). Third, instead of implementing each circuit individually, they found a way to run four circuits in parallel, allowing them to use all 20 qubits in the H1-1. Credit: Adam Malin/ORNL

Using the full capabilities of the Quantinuum H1-1 quantum computer, researchers from the Department of Energy's Oak Ridge National Laboratory not only demonstrated best practices for scientific computing on current quantum systems but also produced an intriguing scientific result.

By modeling singlet fission—in which absorption of a single photon of light by a molecule produces two excited states—the team confirmed that the linear H_4 molecule's energetic levels match the fission process's requirements. The linear H_4 molecule is, simply, a molecule made of four hydrogen atoms arranged in a linear fashion.

A molecule's energetic levels are the energies of each quantum state involved in a phenomenon, such as singlet fission, and how they relate and compare with one another. The fact that the linear molecule's energetic levels are conducive to singlet fission could prove to be useful knowledge in the overall effort to develop more efficient solar panels.

"This is one of the leading motivating factors behind singlet fission—conventional solar cells have a theoretical maximum efficiency of about 33%, but it has been postulated that materials that exhibit singlet fission can break that limit and can be more efficient," said Daniel Claudino, a research scientist in ORNL's Quantum Computational Science group and the project's principal investigator. "The downside is that to understand fundamentally whether a certain material exhibits singlet fission is very hard. There is a specific energetic requirement, and it's difficult to find materials that fulfill it."

With its high accuracy for a manageable computational cost, the ORNL team's approach to using a quantum computer provides an effective simulation method to identify molecules that demonstrate singlet fission properties while bypassing approximations commonly found in techniques used for classical computers. The results of its work were published in *The Journal of Physical Chemistry Letters*.

Singlet fission is a multistate phenomenon, so the ORNL team needed a computational method that could describe all the process's quantum states on an equal footing to calculate accurate energetics numbers. They turned to PDS, which is a quantum solver based on the Peeters-Devreese-Soldatov approach and developed at Pacific Northwest National Laboratory.

PDS holds some advantages over classical strategies for determining a material's energetic properties, including much higher accuracy than density functional theory and fewer computational demands than coupled cluster theory. And, because it was developed to improve the accuracy and efficiency of simulations in quantum chemistry, PDS is well suited to leverage the potential advantages of quantum computers.

"The energetics of singlet fission revolve around double electronic excitations—two electrons move up two higher energy levels simultaneously, which is quite difficult to pin down with algorithms for conventional computers," Claudino said.

"But the underlying way that a quantum computer works, it can naturally treat the quantum correlations that give rise to this singlet-fission phenomenon. That's when we came to the realization that, 'yes, we should use a quantum computer to treat something that is inherently quantum." That's well known. But I think we were the first to realize that it had the application for this specific problem."

Access to the H1-1, an enterprise-ready quantum computer built by Quantinuum (formerly Honeywell), was provided by the Quantum Computing User Program at the Oak Ridge Leadership Computing Facility, a DOE Office of Science user facility.

Quantum computing—a technology still in its formative stages compared to classical supercomputers such as the OLCF's exascale-class Frontier—utilizes quantum bits, or qubits, to perform calculations. Unlike binary bits used in classical computers, qubits go beyond 1s and 0s to also use 1 and 0 simultaneously in a mixed superposition, exponentially increasing its processing power for certain equations, such as those based in quantum mechanics. However, quantum computer systems are still prone to high error rates, and the team had to compensate for this challenge to achieve reliable results.

"It's better to have many more measurements to be on the safe side when it comes to circumventing errors, but then we wouldn't be able to run this algorithm in a timely manner," Claudino said.

"That's when we came up with measurement optimization to bring down the size of our calculations to something that was reasonable in terms of computing time. We went from something that was prohibitively large to something that was amenable to the quantum hardware."

ORNL team members applied three independent strategies to decrease the problem's computational workload, which reduced their time to solution from months to a few weeks. First, in a technique called qubit tapering, they decreased the number of qubits required to express the problem, reducing the size of the problem itself. Second, they took fewer measurements to solve the problem by measuring groups of terms once rather than measuring each individual term from every group. Third, instead of implementing each circuit individually, they found a way to run four circuits in parallel, allowing them to use all 20 qubits in the H1-1.

"We realized that if we wanted to just throw this entire thing into a quantum computer, it's not going to work because it's still too much for the current technology. The idea is that you want to envision a way to tap into the quantum computer but only for specific tasks that we know they can perform better than conventional computers," Claudino said.

"Yet, even then, you're still limited by the current state of the art that only allows us to either go up to a certain size or perform tasks that only take so long. That's the major bottleneck when turning to quantum computers."

The ORNL team's project demonstrated the viability of current quantum computers to tackle scientific problems that could impact daily life. Although Claudino does not foresee tackling singlet fission again soon, his team is considering other problems—such as "the direction of matter and light"—that may be solved using the quantum computing techniques demonstrated in this project.

Even though the approaches we used have been previously published, I would say they are far from being widely adopted. I think we make a strong case for the use of such approaches," Claudino said. "Researchers should be mindful that they may be wasting quantum resources and potentially increasing errors in their simulations by not taking advantage of these techniques."

More information: Daniel Claudino et al, Modeling Singlet Fission on a Quantum Computer, *The Journal of Physical Chemistry Letters* (2023). DOI: 10.1021/acs.jpclett.3c01106

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