Physicists observe rare resonance in molecules for the first time

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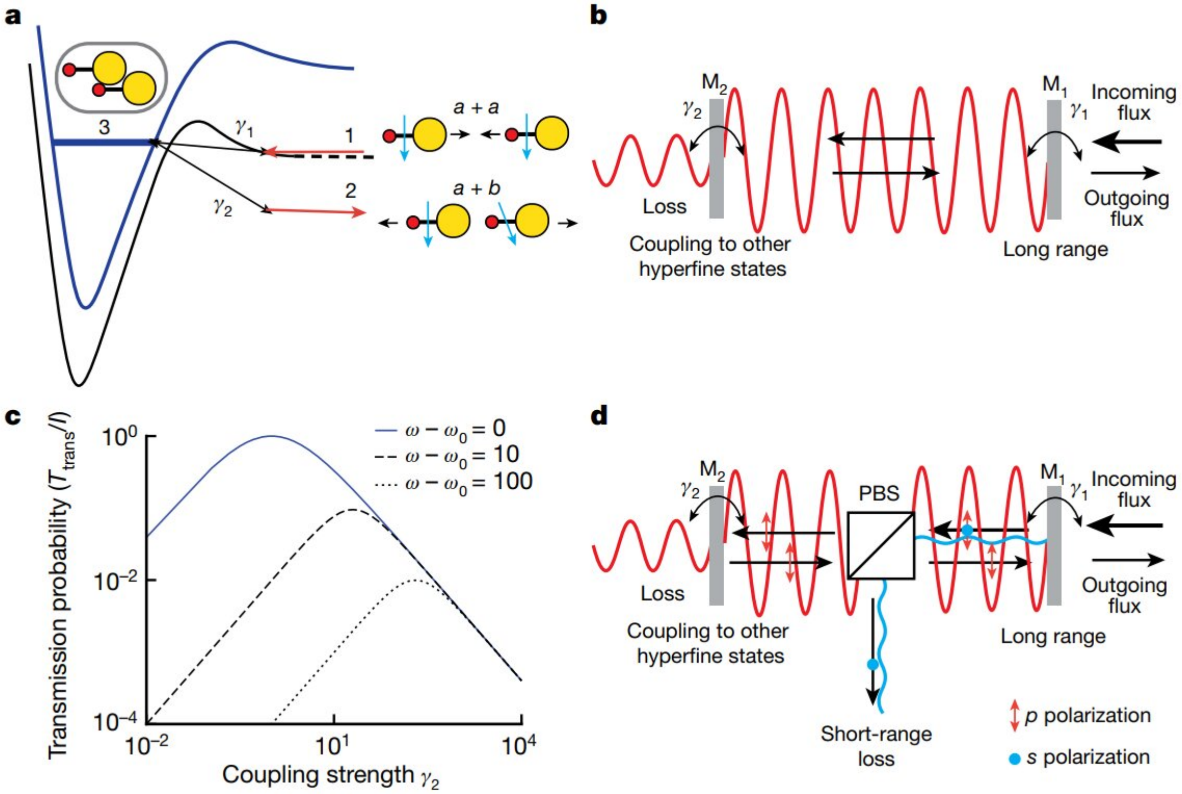

Three-state model for the resonance and optical analogues. Credit: *Nature* (2023). DOI: 10.1038/s41586-022-05635-8

If she hits just the right pitch, a singer can shatter a wine glass. The reason is resonance. While the glass may vibrate slightly in response to most acoustic tones, a pitch that resonates with the material's own natural frequency can send its vibrations into overdrive, causing the glass to shatter.

Resonance also occurs at the much smaller scale of atoms and molecules. When particles chemically react, it's partly due to specific conditions that resonate with particles in a way that drives them to chemically link. But atoms and molecules are constantly in motion, inhabiting a blur of vibrating and rotating states. Picking out the exact resonating state that ultimately triggers molecules to react has been nearly impossible.

MIT physicists may have cracked part of this mystery with a new study appearing in the journal *Nature*. The team reports that they have for the first time observed a resonance in colliding ultracold molecules.

They found that a cloud of super-cooled sodium-lithium (NaLi) molecules disappeared 100 times faster than normal when exposed to a very specific magnetic field. The molecules' rapid disappearance is a sign that the magnetic field tuned the particles into a resonance, driving them to react more quickly than they normally would.

The findings shed light on the mysterious forces that drive molecules to chemically react. They also suggest that scientists could one day harness particles' natural resonances to steer and control certain chemical reactions.

"This is the very first time a resonance between two ultracold molecules has ever been seen," says study author Wolfgang Ketterle, the John D. MacArthur Professor of Physics at MIT. "There were suggestions that molecules are so complicated that they are like a dense forest, where you would not be able to recognize a single resonance. But we found one big tree standing out, by a factor of 100. We observed something completely unexpected."

Ketterle's co-authors include lead author and MIT graduate student Juliana Park, graduate student Yu-Kun Lu, former MIT postdoc Alan Jamison, who is currently at the University of Waterloo, and Timur Tscherbul at the University of Nevada.

A middle mystery

Within a cloud of molecules, collisions occur constantly. Particles may ping off each other like frenetic billiard balls or stick together in a brief yet crucial state known as an "intermediate complex" that then sets off a reaction to transform the particles into a new chemical structure.

"When two molecules collide, most of the time they don't make it to that intermediate state," says Jamison. "But when they're in resonance, the rate of going to that state goes up dramatically."

"The intermediate complex is the mystery behind all of chemistry," Ketterle adds. "Usually, the reactants and the products of a chemical reaction are known, but not how one leads to the other. Knowing something about the resonance of molecules can give us a fingerprint of this mysterious middle state."

Ketterle's group has looked for signs of resonance in atoms and molecules that are super-cooled, to temperatures just above absolute zero. Such ultracold conditions inhibit the particles' random, temperature-driven motion, giving scientists a better chance of recognizing any subtler signs of resonance.

In 1998, Ketterle made the first ever observation of such resonances in ultracold atoms. He observed that, when a very specific magnetic field was applied to super-cooled sodium atoms, the field enhanced the way the atoms scattered off each other, in an effect known as a Feshbach resonance. Since then, he and others have looked for similar resonances in collisions involving both atoms and molecules.

"Molecules are much more complicated than atoms," says Ketterle. "They have so many different vibrational and rotational states. Therefore, it was not clear if molecules would show resonances at all."

Needle in a haystack

Several years ago, Jamison, who at the time was a postdoc in Ketterle's lab, proposed a similar experiment to see whether signs of resonance could be observed in a mixture of atoms and molecules cooled down to a millionth of a degree above absolute zero. By varying an external magnetic field, they found they could indeed pick up several resonances amid sodium atoms and sodium-lithium molecules, which they reported last year.

Then, as the team reports in the current study, graduate student Park took a closer look at the data.

"She discovered that one of those resonances did not involve atoms," Ketterle says. "She blew away the atoms with laser light, and one resonance was still there, very sharp, and only involved molecules."

Park found that the molecules seemed to disappear—a sign that the particles underwent a chemical reaction—much more quickly than they normally would, when they were exposed to a very specific magnetic field.

In their original experiment, Jamison and colleagues applied a magnetic field that they varied over a wide, 1,000-Gaussian range. Park found that molecules of sodium-lithium suddenly disappeared, 100 times faster than normal, within a tiny sliver of this magnetic range, at about 25 milli-Gaussian. That's equivalent to the width of a human hair compared to a meter-long stick.

"It takes careful measurements to find the needle in this haystack," Park says. "But we used a systematic strategy to zoom in on this new resonance."

In the end, the team observed a strong signal that this particular field resonated with the molecules. The effect enhanced the particles' chance of binding in a brief, intermediate complex that then triggered a reaction that made the molecules disappear.

Overall, the discovery provides a deeper understanding of molecular dynamics and chemistry. While the team does not anticipate scientists being able to stimulate resonance, and steer reactions, at the level of organic chemistry, it could one day be possible to do so at the quantum scale.

"One of the main themes of quantum science is studying systems of increasing complexity, especially when quantum control is potentially in the offing," says John Doyle, professor of physics at Harvard University, who was not involved in the group's research. "These kind of resonances, first seen in simple atoms and then more complicated ones, led to amazing advances in atomic physics. Now that this is seen in molecules, we should first understand it in detail, and then let the imagination wander and think what it might be good for, perhaps constructing larger ultracold molecules, perhaps studying interesting states of matter."

More information: Juliana Park, A Feshbach resonance in collisions between triplet ground-state molecules, *Nature* (2023). DOI: 10.1038/s41586-022-05635-8.
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