

QUANTUM TECHNOLOGY

Wave packets get a kick

Intense femtosecond pulses of infrared light can manipulate molecules. It is now shown that such control even extends to making different molecular eigenstates interfere with each other in a way never considered before — a potential tool for optically engineered chemical reactions and for ultrafast information encoding and manipulation.

Thomas Baumert

Quantum mechanics tells us that a classical oscillating point mass can be thought of in terms of a superposition of delocalized eigenmodes of a harmonic oscillator¹ — better known as a wave packet. The advent of ultrashort pulses of light made this temporal aspect of the connection between waves and matter even more important. Laser pulses that are short in comparison with the typical period of a molecular vibration have an intrinsic spectral width that determines the number of quantum-mechanical vibrational eigenstates that couple to the wave packet. The contribution of each eigenstate is time dependent during the excitation and becomes stationary afterwards. In a loss-free environment such wave packets would collapse and revive owing to the anharmonicity in molecular potential², and the population of each eigenstate would remain constant. Now, as they report in *Nature Physics*³, Kenji Ohmori and co-workers from several institutions in Japan have used a cleverly designed experiment to show how a further optical excitation can change this picture and provide a potentially useful tool for taking advantage of molecules in quantum computing.

Ohmori and his colleagues fired femtosecond optical pulses at gas-phase iodine molecules to prepare a vibrational wave packet in the so-called B state (Fig. 1). These molecules then interacted with a strong non-resonant infrared femtosecond pulse — an effect known as impulsive stimulated Raman scattering. This second pulse was delayed by a known time with respect to the excitation pulse. The team monitored the population of individual vibrational eigenstates after both laser interactions using a third laser: a tunable narrowband nanosecond laser, which excites specific vibronic transitions to a higher state known as the E state after a time delay equivalent to several tens of thousands of classical oscillation periods. The fluorescence associated with the decay of this state gives specific information about the population in a vibrational

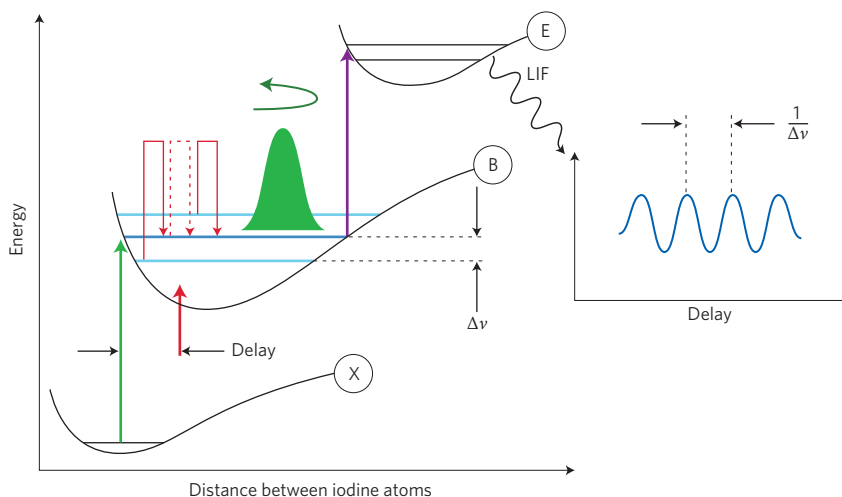


Figure 1 | Schematic of the procedure for making molecular eigenstates interfere. A femtosecond pulse (green arrow) excites a wave packet out of the X state so that it oscillates in the B state of iodine molecules (green). This wave packet then interacts with a time-delayed non-resonant infrared femtosecond pulse (red arrow). The population in each vibrational state (blue lines) is measured using laser-induced fluorescence (LIF) from the E state, which is excited with a narrowband tunable nanosecond laser pulse (purple arrow). As a function of the delay between the two femtosecond pulses, each population oscillates at a rate equal to the difference in frequency between its neighbouring states $\Delta\nu$, however with a different phase. This observation is explained as an interference effect between different possible quantum-mechanical pathways: with the ground state as the common initial state, a specific final vibrational eigenstate in the B state (dark blue) can be reached either by Rayleigh scattering (dashed red curve) or by Raman scattering (solid red curves) from the neighbouring states after the infrared pulse.

mode of the molecule. This enabled the researchers to plot the population in each vibrational state as a function of the delay between the two femtosecond laser pulses. They found that each population oscillates at a rate equal to the frequency difference between its neighbouring states. Moreover, they measured a phase shift between different eigenstates. When the scientists looked at this beating over even longer periods, they found that in fact more than just the next-neighbour frequencies were involved and that the beat shows a similar behaviour to the collapse and revival expected with an unperturbed wave packet.

The authors explain these observations as an interference effect between different

possible quantum-mechanical pathways: with the ground state as the common initial state, a specific final vibrational eigenstate in the B state can be reached either by Rayleigh scattering or by Raman scattering from the neighbouring states after the infrared pulse. They also hint that the different phases in these two processes could be addressed by changing the phase of the infrared pulse, and that in general the mechanism is not limited to vibrational eigenstates.

So is this interference mechanism useful in ways other than being a beautiful picture-book experiment? The answer lies in the identification of research areas in which the interplay of the phases of molecular quantum states and those

of laser fields play a crucial role. Two such fascinating fields can be identified immediately. One is 'coherent control', where the thrilling ultimate goal is to guide chemical reactions by using the tailored coherence properties of laser light^{4,5}. As well as selective bond breaking and bond rearrangements, selective bond formation also has been demonstrated recently⁶ using feedback-optimized laser pulses⁷. Complementarily, experiments under gas-phase collision-free conditions, such as the one presented by Ohmori *et al.*³, could identify individual control mechanisms especially in the promising non-perturbative regime of light-matter interaction. Direct comparisons between theory and such investigations⁸ could shed light onto the 'black-box type' feedback-optimized experiments, and have the potential to increase the efficiency and the selectivity when controlling photochemical systems.

The other possible application is in 'quantum information technology'. Here, combining shaped femtosecond light fields with the excitation of internal motional states of molecules has the exciting prospect of enabling decoherence-free logic operations and algorithms at the speed of molecular dynamics⁹, although several important barriers have to be overcome before this goal can be achieved¹⁰. Considering that silicon-based technology will soon reach its fundamental limits, this experiment³ could also lead to progress in classical information processing as it opens a new route to ultrafast information encoding and manipulation using single-particle interference.

It is the development of sophisticated tools that have traditionally enabled engineers to build instruments that lead us towards new horizons. The same now holds for quantum engineering with the help of tailored light fields. □

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PLANETARY IMAGING

Messages from Mercury

In 2004, NASA launched its Messenger spacecraft. On 17 March this year, following two flybys of Venus and three of Mercury, Messenger became the first mission to enter orbit about Mercury. By the end of the month, it was returning remarkable images of the planet's surface, including this one — the first — dominated by the spectacular Debussy crater.

Named for French composer Claude Debussy, the crater has an impressive system of 'rays', created by ejecta, extending over hundreds of kilometres. Also in the lower sections of the image are areas of the surface that have never been imaged before: NASA's Mariner 10 viewed only about 45% of the planet on flybys in the mid 1970s.

Messenger — an acronym for Mercury surface, space environment, geochemistry and ranging — carries instruments to probe key issues, such as the density and geological history of the planet, and the nature of its magnetic field. Mercury and Earth are the only inner planets to have global magnetic fields, although Mercury's is about 100 times weaker than Earth's, at the planetary surface.

Messenger will remain in orbit for the next year. That's the next Earth year — equivalent to more than four years on Mercury, but only two of its solar days.

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