

Ultrafast physics

Quantum control with a twist

Yaron Silberberg

Laser pulses can be generated such that their shape and state of polarization change on the scale of a few femtoseconds, adding a new twist to the control and manipulation of molecules.

When driving a car, steering is crucial for getting where you want to go. Imagine driving a car with no steering-wheel in Manhattan: you could go to some places, along north–south avenues or east–west streets, but you would be very limited in your choice of destination. Laser scientists faced a similar limitation when trying to drive quantum systems, such as atoms or molecules, into desired states. The linearly polarized laser pulses they used could drive the system along one axis only; full control would require more complex light signals. But a series of advances in ultrafast optics — the latest reported by Suzuki *et al.*¹ and Brixner *et al.*² in *Physical Review Letters* — has now put a steering-wheel in their hands.

When a light pulse is short enough, it interacts with atoms and molecules before they can be affected by their environment. The interaction is then described by simple quantum mechanical rules. By carefully tailoring the shape of the optical pulse, it is possible to manoeuvre the system into desirable final states, particularly those that are hard to reach through simple thermodynamic processes. For example, it might be possible to break a certain bond in a molecule while leaving other, perhaps weaker,

bonds intact. The general approach is known as quantum coherent control, a field that developed as a theoretical exercise in the mid-1980s, but which has seen intense experimental effort in recent years³.

Coherent-control experiments start with light pulses that last typically a few tens of femtoseconds (one femtosecond is 10^{-15} seconds). Such pulses are now routinely produced by commercial lasers. The pulses are sent through an optical set-up known as a pulse shaper, which can be programmed to generate complex temporal shapes. The shaper acts as a frequency-domain synthesizer, separating a short pulse into many frequency components. The phase, and possibly the amplitude, of each component can be tweaked individually. The result is a longer pulse with an internal structure that can be defined with great precision.

It used to be the case that all quantum control experiments with shaped pulses used linearly polarized light — light whose electric-field vector is confined to a single direction. The optical field of a linearly polarized pulse puts a force on the charges in the system — be they electrons or ions — along only one direction. Now, it is quite easy to convert this linearly polarized light into

other polarization states, say circular or elliptical ones, by placing simple polarization converters in the beam. Yet this modifies the polarization of the entire pulse uniformly. Stretching the analogy made earlier, this is like driving a car whose steering-wheel is stuck, forcing it to turn constantly to one side — not a much better situation.

What is needed is the ability to change the driving direction continuously — that is, to modify the polarization direction within the optical field. This was recently realized by Brixner and Gerber⁴: in their polarization pulse shaper, not only the amplitude and phase but also the polarization state of the different frequency components can be changed, creating pulses with complex, twisted polarization structures. In such a pulse, the polarization direction may change on the scale of a few femtoseconds.

The polarization shaper was first used to perform a relatively simple task. By rotating the polarization of a narrow band of frequencies within the broader pulse spectrum, my group⁵ was able to generate two synchronized pulses whose polarization was at right angles to each other. This was used in a technique for nonlinear molecular spectroscopy known as CARS, which usually requires two laser sources. However, the main interest in polarization-shaped pulses stems from their ability to impose and control rotations. Light pulses with varying polarizations can transfer angular momentum and therefore either drive a system into a final rotating state or use rotational states as intermediates in more complex interactions. Both goals have been demonstrated in atomic and molecular systems.

In atoms, because of their symmetry, the situation is somewhat simpler than it is in molecules. Coherent control of the angular-momentum states of atoms using polarization-shaped pulses has been demonstrated by Dudovich *et al.*⁶: we showed that careful crafting of the polarization and phase of the pulse can excite, via multi-photon absorption, particular states that cannot be recognized by linearly polarized control. Controlling angular momentum in molecules is a greater experimental challenge. To start with, although molecules have preferred directions, in most experimental situations they are oriented randomly in space. In addition, the energy-level structure that is quite simple in atoms becomes more complex, as electronic states are combined with vibrational and rotational molecular states. This complexity demands more elaborate schemes for control that cannot rely on perfect knowledge of the system at hand.

Now the potential of polarization shaping for controlling molecular processes has at last been demonstrated. Suzuki *et al.*¹ and Brixner *et al.*² have investigated the ionization of diatomic molecules that were pre-aligned (to

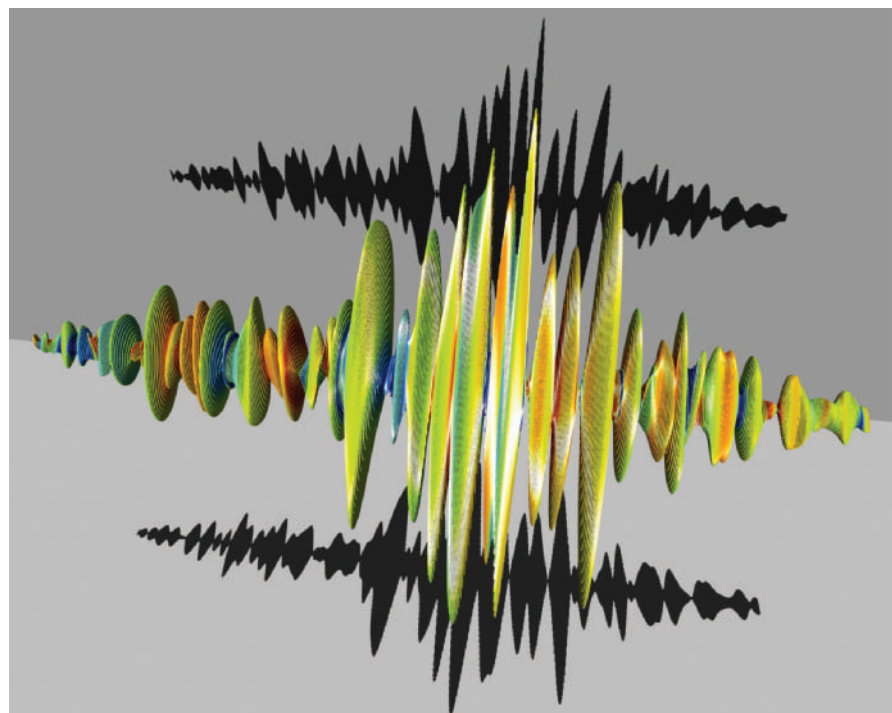


Figure 1 Polarization-shaped pulse, optimized for the ionization of potassium molecules². Ellipses represent the amplitude of the electric field; colours indicate different frequencies.

make sure they respond uniformly) and then irradiated with polarization-shaped pulses. These molecules — iodine¹ and potassium² — are as simple as molecules can be, but they are already too complex to theoretically design an optimized polarization structure that would maximize ionization. Therefore, both experiments relied on self-learning techniques, in which the optimal pulse shape and polarization structure (Fig. 1) are found through an iterative optimization procedure. Both groups have shown conclusively that pulses with complex polarization structures ionize these molecules more efficiently than pulses with a uniform polarization.

Further work will surely follow, using polarization-shaped pulses to tweak atomic and molecular systems with greater precision. Several proposals have already been

made and await experimental tests, including the alignment of molecules in a gas phase, the manipulation of chiral molecules and the control of attosecond pulse formation. Expect laser scientists to steer towards even more uses for these twisted pulses of light. ■

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Genomes

Worming into genetic instability

Susan M. Rosenberg and P. J. Hastings

A study of roundworms shows that genomic mutations occur surprisingly frequently, and that the kinds of changes involved differ from those predicted. Are genomes inherently less stable than previously suspected?

DNA carries the coded information that specifies the size, shape, body plan and many other basic characteristics of most organisms. To transmit these characteristics faithfully, DNA must pass from generation to generation with relatively few mutations. But mutations do happen, and can have profound consequences. These include inherited diseases, cancer and drug-resistant infections, but also the genetic differences among individuals that, through natural selection, drive evolution.

Until now, mutations seemed to be relatively rare and to occur in a characteristic spectrum. But such observations are challenged in the paper by Denver and colleagues on page 679 of this issue¹. These authors used a particularly powerful way to hunt for mutations in the roundworm *Caenorhabditis elegans* (Fig. 1, overleaf) — and found at least ten times more mutations, and a different assortment, than anticipated.

Traditional ways of estimating mutations are indirect², involving either phylogenetic studies of wild organisms or phenotypic methods in the lab. Phylogenetic studies involve comparing DNA sequences between species and estimating the number and kinds of changes that have occurred since the species diverged. Phenotypic methods rely on the ability of some mutations to change a trait (phenotype) of an organism. After a defined number of generations, rare mutants carrying the new trait are quantified, and mutation rates are calculated and then extrapolated to predict rates for the whole

genome. This extrapolation takes into account the genome's size and the fraction of mutations that has been estimated to produce phenotypic change (about one-third)².

However, both approaches probably underestimate the inherent mutation rate and skew the variety of mutations found. For instance, some mutations are harmful, and so the organisms that carry them are less likely to contribute to the next generation (they are 'selected against'), both in the wild and in large cultures. And the fraction of mutations that produces no phenotypic change might be larger than imagined.

Denver *et al.*¹ bypassed the phenotype-bias problem by directly sequencing randomly chosen stretches of DNA in laboratory-grown worms. They also minimized selection against harmful mutations by maintaining many lines of worms, separating a single worm from each progeny and allowing it to produce the next generation by self-fertilization, without competing with other worms. Rapid and severe loss of fitness occurs in these worms because, when their numbers are reduced to one repeatedly, random mutations become fixed — a phenomenon known as Muller's ratchet³.

From these pampered worms, Denver *et al.* sequenced four million base pairs of DNA, and found 30 new mutations compared with the original animals. This equates to a rate of 2.1 mutations per genome per generation. This rate is at least ten times higher than those reported previously in worms and other DNA-based organisms,



100 YEARS AGO

Prof. Schäfer, F.R.S., describes a simple and efficient method of performing artificial respiration in the human subject, especially in cases of drowning... Immediately the patient is recovered from the water he is placed face downwards, the head being turned sideways so that the mouth and nose are unobstructed, with a folded coat under the lower part of the chest; if respiration has ceased every instant of delay is serious. The operator then places himself athwart, or on one side of, the patient's body in a kneeling posture and facing the head. He places his hands flat over the lower part of the back (on the lowest ribs), one on each side, and gradually throws the weight of his body on to them so as to produce firm pressure — which must not be violent — on the patient's chest. This compresses the chest, and air (and water if there be any) is driven out of the patient's lungs. He then raises his body slowly so as to remove the pressure, still keeping his hands in position. This process of applying pressure and of relaxation of pressure by the forward and backward movement of the operator's body is repeated every four or five seconds without any marked pause between the movements. This course must be pursued for at least half an hour, or until the natural respirations are resumed... If there be means, others may remove the wet clothing by cutting it off, and may apply hot flannels to the body and limbs and hot bottles to the feet.

From *Nature* 4 August 1904.

50 YEARS AGO

The United Kingdom Atomic Energy Authority has announced that a heavy-water reactor (or atomic pile) which has been built at the Atomic Energy Research Establishment, Harwell, is now in operation. 'Dimple' is a low-powered thermal neutron research reactor. The heavy-water moderator is contained in a tank which is surrounded by a graphite neutron reflector. Outside this is a concrete radiation shield. The reactor fuel is submerged in the heavy water. Both the type of fuel and its arrangement in the tank can be changed quickly so that what is, in effect, a different design of reactor can be built up in a matter of days... The versatility of Dimple will make it an extremely valuable tool in the design of future power-producing reactors and for measuring essential constants in reactor physics.

From *Nature* 7 August 1954.