MOLECULAR PHYSICS

Subradiance spectroscopy

Subradiant states have remained elusive since their prediction sixty years ago, but they have now been uncovered in ultracold molecules, where they could prove useful for ultra-high precision spectroscopy.

Benjamin Pasquiou

For many decades molecular spectroscopy has provided deep insights into the structure and properties of matter. As they report in *Nature Physics*, Bart McGuyer and colleagues[1] now demonstrate a significant step forward for spectroscopy with ultracold molecules. By adapting atomic clock techniques, they characterized the optical transition oscillator strengths of subradiant molecular states with exquisite precision. They then explored the fundamental effect of cooperative light emission, which could lead to further improvements in ultra-high resolution molecular spectroscopy.

The concepts of superradiance and subradiance were first described in a quantum mechanical framework in a paper published in 1954 by Dicke[2]. Atoms within a dilute ensemble initially prepared in an excited electronic state spontaneously emit photons independently (Fig. 1a). This gives rise to an exponential decay in the number of emitted photons with a rate Γ determined by the electronic structure of the atom. But, if the distance between atoms becomes small compared with the wavelength of the radiation field, the atomic dipoles will experience phase locking and exhibit cooperative emission. This emission is strongly dependent on the symmetry of the ensemble state of correlated atoms. For superradiant states (Fig. 1b), the emission may be amplified to intensities proportional to N^2 instead of N, where N is the number of emitters, with an accelerated decay rate up to $N \times \Gamma$. In contrast, for subradiant states (Fig. 1c), the decay rate approaches zero, giving rise to a spectroscopic feature with an ultranarrow linewidth.

Whereas superradiance has been intensively studied both theoretically and experimentally[3,4], studies of subradiance have remained scarce[5]. This is in large part due to the intrinsic low emission rate of subradiant states and their fragility to spurious effects such as atomic motion (Doppler effect), atomic interactions and the existence of other decay paths. Creating systems with a distance between atoms smaller than the radiation wavelength poses yet another challenge[6]. Homonuclear molecular dimers have excited states with a superradiant or subradiant character, depending on whether the central symmetry of the electronic cloud has the same or opposite parity as that of the molecular ground state. Superradiance and subradiance are, in this case, restricted to the simplest, yet physically meaningful, case of two cooperating atoms.

McGuyer *et al.*[1] demonstrate direct measurement and in-depth characterization of the properties of subradiant strontium dimers with near-dissociation energies. Their achievement relies on several techniques. First, they trap and laser cool ^{88}Sr atoms down to temperature of around 1 μK and then associate pairs of atoms into molecules in the $X^1\Sigma_g^+$ molecular ground state using an optical transition — photoassociation. Despite these low temperatures, the residual motion of the atoms would still significantly broaden spectroscopy signals, due to the Doppler effect, thus masking features as narrow as a subradiant transition linewidth. To get rid of these effects, McGuyer and co-workers keep the molecules in the tight-binding regime inside a one-dimensional optical lattice — a standing wave that traps molecules in a collection of pancake-like sites. Unfortunately, the addition of this external electric field can affect the energy of the molecular states being studied. This usually leads to systematic shifts of the transition frequency and to the broadening of spectra due to the variation of the laser intensity across the spatial extent of the molecular cloud.

To access the unperturbed properties of the subradiant states, the authors have adapted a technique used for atomic clocks. By properly choosing the wavelength and polarization of the light, they engineer a so-called magic optical lattice that affects both ground and excited molecular states in exactly the same way. Combining this lattice with a narrow linewidth (<200 Hz) probe laser and good control over the external magnetic field, they successfully probed several subradiant states of the 1_g molecular potential (corresponding asymptotically to $^1S_0 + {}^3P_1$ at long interatomic distances). They measured the oscillator strengths of optical transitions towards these states, and measured linewidths as small as 30 Hz — almost 300 times narrower than the atomic (non-cooperative) linewidth.

A similar previous study[7] was able to determine the binding energy of several states in homonuclear ytterbium dimers and to give evidence of their subradiant nature. What is truly remarkable in the present work is that the precision achieved now

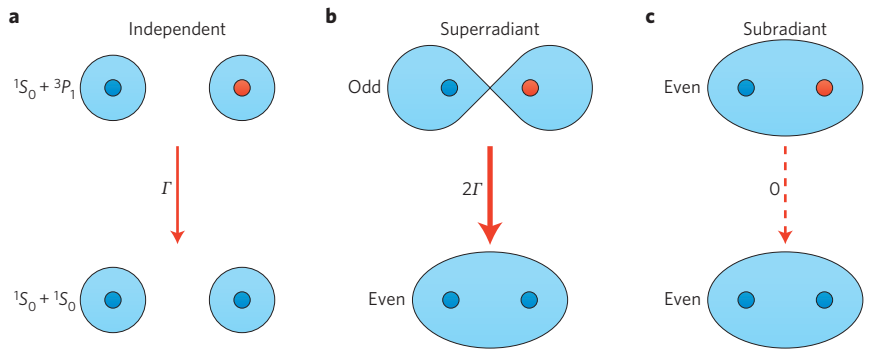

Figure 1 | Cooperative and independent spontaneous emission processes. **a**, The spontaneous emission from an independent atom decaying from an excited state (red dot) towards the ground state (blue dot) occurs at a rate Γ, which is determined by the electronic structure of the atom. **b**, The cooperative emission of a superradiant molecular dimer ($N = 2$, two emitters) with opposite electronic orbital symmetry compared with that of the ground state, has an enhanced decay rate of 2Γ. **c**, McGuyer *et al.*[1] use ultra-high precision spectroscopy to study subradiant molecular states of $^{88}Sr_2$, which should feature a zero emission rate.

allows McGuyer and co-workers to probe inherent molecular phenomena that increase the linewidth of subradiant transitions to finite values. The authors relate these perturbations to two effects: radiative decay via the higher-order magnetic dipole and electric quadrupole transitions, and non-radiative decay via non-adiabatic Coriolis mixing towards the $^1S_0 + {}^3P_0$ lower-lying continuum. They theoretically account for the respective contributions of these two effects and find satisfying agreement with experiment. Interestingly, they also demonstrate coherent manipulation of subradiant states by inducing Rabi oscillations.

All in all, the demonstration of the tremendous precision achievable by spectroscopy of ultracold molecules promises a wealth of future measurements exploiting the complex structure of these systems. Foreseeable applications range from benchmarking of *ab initio* quantum chemistry calculations, to testing for variation of fundamental constants[8]. ❐

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TEN YEARS OF NATURE PHYSICS

Slowly but surely

In 2006, *Nature Physics* published a paper reporting a Stern–Gerlach effect for dark polaritons and one revealing the existence of slow-light solitons. Both of these papers have significantly advanced the field of slow-light research.

Ebrahim Karimi and Robert W. Boyd

Slow-light research[1] explores ways of dramatically slowing down the speed of light pulses travelling through an optical medium. Two papers published in *Nature Physics* in 2006 significantly advanced this field, revealing unexpected effects. Leon Karpa and Martin Weizt reported the observation of a Stern–Gerlach effect for dark polaritons propagating through a rubidium vapour under slow-light conditions created by electromagnetically induced transparency[2]. And Joe Mok and colleagues observed a slow-light optical soliton propagating in a shape-invariant manner through a nonlinear dispersive material[3].

The concept of velocity is well-defined for particles, but it is much murkier for waves. In fact, as a consequence of dispersion, a pulsed beam spreads and is distorted during propagation. Therefore, a unique value of the velocity cannot be assigned to a pulse propagating inside a material medium. This is due to the fact that a pulse comprises a superposition of an infinite number of monochromatic waves. These waves interfere either constructively or destructively in such a way that the beam is both spatially and temporally well localized.

When such a pulse traverses a dispersive medium, the relative phases among the various spectral components of the pulse change during propagation. Thus, it causes a change to the effects of destructive and constructive interference, consequently modifying the position (and shape) of the pulse compared with propagation in vacuum. The velocity with which a pulse moves though such a medium is often well described by the group velocity, which is dependent on a dispersive term that can be manipulated to slow the light pulse down to everyday human velocities. There are two ways to achieve this control[4]: by using the dispersive term in the group index of the material medium, or by exploiting structural resonances. The two *Nature Physics* publications[2,3] each report a different approach.

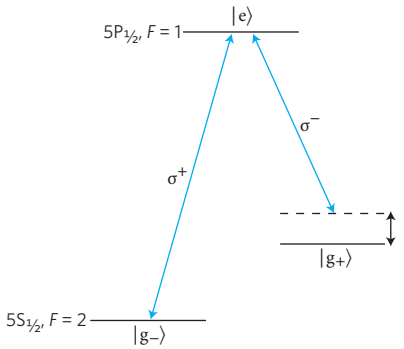

Figure 1 | Simplified rubidium energy-level diagram showing the conditions for establishing electromagnetically induced transparency and producing a strong slow-light effect. The destructive interference between the two pathways (defined by the polarization of the light; either σ^+ or σ^-) leading to the excitation of the upper level creates a dark polariton — a quasiparticle in which the photon and atomic excitation are strongly coupled. Because of the strong coupling, a photon acquires atomic-like properties, making a Stern–Gerlach effect possible. Reproduced from ref. 2.

In the original experiment by Stern and Gerlach, a beam of silver atoms passed through a region of non-uniform magnetic field. Each atom then experienced a force and, as the silver atoms had zero orbital angular momentum, the only contribution to the total angular momentum came from electron spin. This resulted in the beam of silver atoms splitting into two components, separated along the direction of the magnetic field gradient. In fact, the Stern–Gerlach experiment was later recognized as the first experimental demonstration of electron spin.

One would not expect photons to exhibit such an effect, because the photon does not possess a magnetic moment — as is the case for all particles with zero rest mass. However, under slow-light conditions, the photon is strongly coupled to the material medium. The experiment by Karpa and Weizt made use of the strong dispersion of atomic rubidium vapour under conditions of slow-light based on electromagnetically induced transparency[5,6] (Fig. 1).

Under these conditions, it helps to think of photons and atoms not as separate entities but rather as a coupled system in the form of a quasiparticle known as a dark polariton[7]. Such a quasiparticle can possess a magnetic moment through the