Coupling Single Molecules to Laser Fields

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Optical studies of single molecules in condensed matter eliminate broadening from static disorder and reveal temporal fluctuations in complex media. Single molecule methods are currently applied to many systems at ambient conditions, to solve problems ranging from physical chemistry to biology [1]. At cryogenic temperatures, single molecules embedded in suitable matrices present narrow homogeneous lines, comparable to those found in atomic physics. These sharp lines lead to large resonant absorption cross-sections, which makes the detection and imaging of single molecules easier than at room temperature. Various properties of single molecules at low temperatures have been investigated in the past ten years: intramolecular dynamics, spontaneous or photo-induced frequency jumps arising from conformational changes in the environment [2]. Here, we concentrate on the coupling of single molecules to light waves for basic experiments in nonlinear and quantum optics.

We have investigated various resonant nonlinear effects with single molecules. In all these experiments, the molecule behaves like an optical two-level system coupled to the radiation field, and its dynamics is quantitatively described by optical Bloch equations. Such simple nonlinear optical effects as optical saturation, or the light-shift of the optical transition by a pump beam would be difficult or impossible to observe on ensembles in condensed matter, but they can be observed easily on single molecules. We also measured more complex effects, such as three-photon (hyper-Raman line), or multi-photon resonances (subharmonic oscillations) in a near-resonant bichromatic optical field. A single molecule can mix a visible frequency with RF frequencies, giving rise to Rabi resonances. In a more recent experiment [3], we proposed to use a single molecule as a triggered source of single photons. The strong coupling of the molecule to an RF field was used to sweep the transition frequency, and to coherently drive the molecule from its ground to its excited state, by adiabatic following. Again, optical Bloch equations are a very reliable guide to determine the distribution of the number of emitted photons emitted upon each passage through resonance. We obtained more than 70 % of passages producing one and only one photon. This single molecule source is much simpler than semiconductor devices based on Coulomb blockade, which so far must operate at mK temperatures [4]. Simpler sources still could be obtained with single molecules at room temperature, with pulsed excitation. Narrow emission lines could be coupled to resonant cavities, to influence the spectrum or directivity of the fluorescence.

As these few experiments show, single molecules in condensed matter can be used as predictable and controllable elements in their interaction with laser fields. The next challenge is to combine several single molecules and resonant elements such as optical cavities to design new experiments in nonlinear and quantum optics.

[1] see the special issue of *Science* **283** (12 March 1999).
[2] Ph. Tamarat et al., *J. Phys. Chem.* **104** 1 (2000).
[3] Ch. Brunel et al., *Phys. Rev. Lett.* **83** 2722 (1999).
[4] J. Kim et al., *Nature* **397** 500 (1999).