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Looking back on 28 years of cryogenic single-molecule experiments

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Abstract. Starting with the first single-molecule fluorescence experiments in 1990, the field of cryogenic single-molecule spectroscopy exploits the narrow zero-phonon lines of single molecules, usually in molecular crystals and glasses. Occasionally, similar experiments can also be done at room temperature, as illustrated by the case of the NV^- center in diamond. In this review contribution, I shall illustrate the variety and scope of the experiments performed in the past 28 years, highlighting some important points and outlooks.

1 Spectroscopy and magnetic resonance

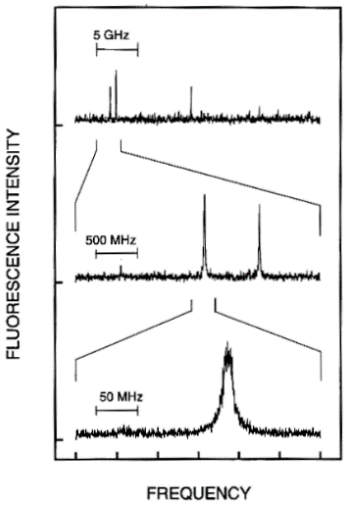

Fig. 1. Example of a single-molecule fluorescence excitation spectrum showing the lifetime-limited lines of single pentacene molecules in a p-terphenyl crystal at 1.8 K (from Ref. 1).

From the first high-resolution optical experiments on single molecules [1], it was clear that this fine spectroscopy enables accurate mapping of molecular electronic and vibronic states through excitation spectra and molecular vibrations from fluorescence spectra. Blinking single-molecule fluorescence yields information about triplet population and depopulation rates, and optically detected magnetic resonance provides the positions of the triplet sublevels split by magnetic spin-spin and spin-orbit interactions [2]. Later work identified hyperfine interactions and achieved magnetic resonance experiments of single nuclear spins through ENDOR [3].

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2 Quantum optics

The narrow zero-phonon lines are often lifetime-limited, which opens unique opportunities for quantum optics. Antibunching in the emission of single molecules was demonstrated early on at low temperature [4]. Single-photon emission at low temperature [5] provides indistinguishable photons, that have been used in Sandoghdar's group for pioneering quantum optics experiments in solid-state phases for the first time, before the experiments with self-assembled quantum dots. Today, single molecules at low temperature are still attractive contenders for single-photon sources delivering indistinguishable photons.

3 External field effects and local probing

Being ultra-narrow oscillators, single molecules are very sensitive probes for all kinds of perturbations, by pressure or mechanical strain, electric fields, magnetic fields. This sensitivity can be exploited to detect small changes in the surroundings of the molecules, for example from acoustic or mechanical deformation [6] or from charge carriers diffusing or trapped in the matrix.

4 Spectral diffusion

The time-dependent dynamics of single-molecule lines has clarified the concepts of spectral diffusion in molecular solids, glasses and polymers at low temperature, by displaying individual two-level systems, and quasi-local vibration modes [7]. Many other causes of spectral diffusion have been identified or proposed in more specific systems, such as phenyl ring flips in p-terphenyl crystals, or rotational-translational coupling in methylated matrixes.

In conclusion, the knowledge and experience accumulated in the previous two decades have made single organic molecules uniquely attractive quantum emitters. Along with self-assemble quantum dots and color centers in diamond, these fascinating systems are excellent candidates for a broad variety of nanophysics experiments, including optomechanics, localized probing, or as the cores of devices for the storage and treatment of quantum information.

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