Single atoms on an optical nanofibre

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Abstract. We show that single atoms can be detected using a sub-wavelength-diameter silica-fibre, an optical nanofibre, and the fact that single photons spontaneously emitted from the atoms can be readily guided into a single-mode optical fibre. Moreover, we find that single atoms around the nanofibre reveal a very prominent spectral feature, that is, the excitation spectrum splits into two peaks with a very small separation, smaller than the natural linewidth.

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1. Introduction

In recent years, significant progress has been achieved in manipulating single atoms, especially towards the development of quantum information technologies. Many kinds of single-atom-localizing and trapping schemes have been demonstrated so far using various external fields. In this context, a major problem arises as to how to detect the small number of atoms. Quite often, these experiments rely on complicated designs of high-numerical aperture optics [1, 2] or high-finesse optical cavities [3, 4] that may add to the technical difficulties. Hence, novel ideas to manipulate and detect single atoms are needed to accelerate the technological advancements. Recent advances in quantum plasmonics, demonstrated for quantum dots, would be a typical example on that line [5, 6].

One promising idea to manipulate a small number of atoms is to use thin optical fibres with sub-wavelength diameter, termed optical nanofibres [7]. It has been theoretically predicted that due to the confinement of the field in the guided mode, spontaneous emission of atoms can be strongly enhanced around the nanofibre and an appreciable amount of fluorescence photons can be channelled into a single-guided mode of the nanofibre [8]. Based on this idea, efficient probing of few atom fluorescence through the guided mode of the nanofibre has been experimentally demonstrated [9]. Moreover, the strong evanescent field around the nanofibre can be efficiently used for trapping, detecting and manipulating atoms. Various atom-trapping schemes have been proposed based on this idea [10, 11]. A novel idea for cavity quantum electrodynamics has also been proposed based on the field enhancement in micro-structured optical fibres [12]. Also, recently, evanescent field absorption spectroscopy of a few atoms around the nanofibre have been experimentally investigated [13]. Furthermore, it has been theoretically demonstrated that two distant atoms on the nanofibre surface can be entangled through the guided mode [14]. Hence, the optical nanofibres can also open new approaches for manipulating single atoms and photons and can prove to be a promising tool in quantum information technology. However, to date there is no experimental work demonstrating single-atom detection or manipulation using optical nanofibres.

In the present paper, we show that single atoms can be detected using an optical nanofibre, and that single photons spontaneously emitted from the atoms can be readily guided into a single-mode optical fibre. Moreover, we find that single atoms around the nanofibre reveal a very prominent spectral feature, that is, the excitation spectrum splits into two peaks with a very small separation, smaller than the natural linewidth. We anticipate that the present results may bridge single-atom physics and optical-fibre technologies, and may give a new perspective for quantum-information technologies.

2. Experimental set-up

Experiments are performed by overlapping cold Cs-atoms in a magneto-optical trap (MOT) with an optical nanofibre and by observing fluorescence photons through the nanofibre after switching off the MOT laser beams. Figure 1 shows the conceptual diagram of the work. The nanofibre is located at the waist of a tapered optical fibre which is produced by heating and pulling commercial single-mode optical fibres. The fibres are adiabatically tapered so that the single-mode propagation condition can be maintained for the whole fibre length. In the present experiments, we use nanofibres [9] with a diameter of 400 nm and a length of 2 mm. An MOT equipped with a resistively heated alkali dispenser source is used to produce cold Cs-atoms.
The MOT position is controlled to overlap with the nanofibre [9]. Atom number density in MOT and MOT size are controlled by adjusting the dispenser current. The maximum number density around the nanofibre is $7 \times 10^9$ cm$^{-3}$ with an MOT diameter of 2 mm and the temperature of atoms around 100 $\mu$K. The MOT laser beams are switched off for 10 $\mu$s periodically at an interval of 200 $\mu$s. During the switched-off periods, atoms around the nanofibre are excited by a probe laser beam, irradiated perpendicularly to the nanofibre in a standing-wave configuration with a polarization perpendicular to the fibre. Fluorescence photons are observed at one end of the fibre using an avalanche photodiode (APD) and a single-photon counting system, and the photon counts are accumulated for many cycles.

3. Results

3.1. Fluorescence excitation spectra: effect of violet laser irradiation

Figure 2 shows the excitation spectra measured for a closed-cycle transition, $6S_{1/2} F = 4 \leftrightarrow 6P_{3/2} F' = 5$, by scanning the probe laser frequency around the atomic resonance. The MOT is set to the maximum density condition and the probe laser diameter is set to 2 mm to irradiate the whole atom cloud. The intensity of the probe laser is set to 3.3 mW cm$^{-2}$. One must note that the intensity is higher than the saturation intensity for the atomic transition, but the spectral profile does not change from that observed for weaker intensities [9]. During the experiments, we found that the excitation spectrum changes drastically when we irradiate the nanofibre with violet laser radiation of wavelength 407 nm. The trace A denotes the observed spectrum before the violet laser irradiation. As reported previously, the observed line shape is quite different from the usual atomic line shape. The spectrum consists of a sharp peak near the resonance and a long tail on the red detuned side. The trace B denotes the observed spectrum after the violet laser irradiation. The conditions are the following: the nanofibre region is irradiated with the violet laser in the presence of the MOT for several minutes. The irradiating power is 5 mW with a beam diameter of 2 mm (the irradiating intensity is 150 mW cm$^{-2}$). After switching-off the violet laser, the fluorescence excitation spectrum is measured. As seen readily, the spectrum is very different from trace A. The spectrum becomes very much squeezed towards the atomic resonance, and the peak fluorescence count increases almost 30 times. The width of the spectrum is 15 MHz FWHM. We should note that irradiation in the absence of the MOT has no effect on the spectrum. We have also found that once the nanofibre is irradiated, the observed effect lasts for several days.
Figure 2. Fluorescence excitation spectra measured through the nanofibre for the closed-cycle transition, $6S_{1/2} F = 4 \leftrightarrow 6P_{3/2} F' = 5$. Detuning is measured with respect to the atomic resonance. Traces A and B correspond to without and with the effect of violet laser irradiation, respectively. The spectra are displayed in two vertical scales so that the change of the spectrum is readily seen.

3.2. Photon correlations

In order to clarify the single-atom characteristics around the nanofibre, we perform Hanbury-Brown and Twiss (HBT) experiments by reducing the atom number. The fluorescence light through the fibre is split into two using a 3 dB fibre coupler, and is detected by two separate APDs. The photon correlations between the two channels are measured in single start-stop mode using a time-correlated photon-counter (TimeHarp-200, PicoQuant GmbH) with a time resolution of 1 ns. In the measurements, the dispenser current is decreased to reduce both the atom number and the MOT size. The minimum MOT size is $80 \mu m$ in diameter with an atom density of $\sim 0.7 \times 10^9$ cm$^{-3}$ for dispenser current $I_D = 3.8$ A. The probe laser is line-focused perpendicularly to the fibre axis, down to $100 \mu m$ using a cylindrical lens to spatially restrict the observation region to the atom cloud, and the frequency is tuned close to the atomic resonance within a few megahertz to maximize the photon count rate. The focused probe laser intensity is $60$ mW cm$^{-2}$. Average atom number in the observation region is estimated to be much less than one for $I_D = 3.8$ A, assuming an observation volume around the nanofibre of 200 nm in thickness [9] and 100 $\mu m$ in length.

The measurements were carried out for both conditions before and after the violet laser irradiation. Single-atom behaviours were clearly observed after the irradiation, but no single-atom behaviour could be observed before the irradiation. In the following, we describe the results obtained for the condition after the irradiation.

The solid curves in figure 3(a) display the observed coincidences for different delay times between the two channels up to $\pm 100$ ns. Each curve is obtained after an integration time of 3 min which requires a measurement time of 1 h. The coincidences for $I_D = 0$ A correspond to the background for the uncorrelated scattered light from the nanofibre. The coincidences for
Figure 3. (a) Photon correlations with HBT arrangement for atoms around the nanofibre. $\tau$ and $I_D$ denote the delay time between the two channels and the dispenser current, respectively. Dashed curves denote the theoretically calculated photon coincidences for $n \sim 0.3$ and $1.7$, respectively, where $n$ denotes the average atom number. (b) The green solid curve shows the normalized correlation for longer timescale, measured through the nanofibre, for $I_D = 3.8$ A. The black dashed curve is the exponential fit to the data giving a decay time of 1.8 $\mu$s.

$I_D = 3.8$ and 4 A show the correlations for the fluorescence photons. Both curves show clear antibunching of fluorescence photons at zero time-delay and Rabi oscillation behaviours in the wings.

The photon coincidences from $N$-atoms is proportional to $Ng^{(2)}(\tau) + N(N - 1)$, where $g^{(2)}(\tau)$ is the correlation function for a single atom [1]$^2$. Since the atom number may fluctuate in the present experiment, we must average over the atom number. As a first approximation, we assume a Poissonian distribution of atom number and hence the observed coincidences are proportional to $g_n^{(2)}(\tau) = \frac{1}{n}g^{(2)}(\tau) + 1$, where $n$ is the average atom number and $g_n^{(2)}(\tau)$ is the normalized correlation function [15]. The observations are fitted assuming spontaneous emission time of 30 ns and Rabi frequency of 13 MHz. The fitting leads to the average atom number $n \sim 0.3$ and $1.7$ for $I_D = 3.8$ A and 4 A, respectively. We should note that the assumed Rabi frequency is 1.3 times smaller in value than that simply estimated for the $F = 4 \leftrightarrow 5$ transition.

Photon coincidences for longer timescale up to 4 $\mu$s are measured for $I_D = 3.8$ A. The normalized coincidences are plotted in figure 3(b) without subtracting the background. The plot clearly shows an exponential decay with a time constant of 1.8 $\mu$s. Fluorescence photon count under this condition is $\sim 1 \times 10^4$ counts s$^{-1}$. So, it should be noted that the single start–stop measurement under the present condition gives a correct temporal behaviour without the piling-up effect [16], since the average photon number for a period of 4 $\mu$s is estimated to be 0.04, much less than one.

$^2$ Note that this formula is not exactly valid under the present single-mode observation conditions, but it is approximately valid for a small number of atoms. Details on this point will be discussed elsewhere theoretically and experimentally.
3.3. Single-atom excitation spectra

We measure the excitation spectra under the single-atom condition, $I_0 = 3.8 \text{ A}$, to further clarify the behaviours of atoms. Observed results for three different probe laser intensities are displayed in figures 4(a)–(c) by solid curves. Measurements are performed by using similar procedures as those for figure 2. Peculiar features are readily seen. For the lowest laser intensity, the spectrum exhibits almost a Lorentzian shape with 8 MHz FWHM, slightly broader than the spontaneous-emission lifetime broadening, but a sharp small dip is seen at the centre. With increasing laser intensity, the signal becomes stronger and the sharp dip becomes more apparent, but the dip width is narrower than the lifetime broadening. We have checked the dependence for the probe laser polarization, but the spectrum does not show any dependence for linear or circular polarization.
4. Discussion

The spectrum with a red tail observed in the trace A of figure 2 is attributed to the van der Waals (vdW) interaction between the Cs-atom and the nanofibre surface, which would be dominant for distances closer than $\lambda/2\pi$ from the surface [9, 17, 18]. It may be understood through a process in which atoms close to the nanofibre fall into a deep vdW potential. The drastic change of the spectrum suggests that the surface of the nanofibre might be modified after the violet laser irradiation, in such a way that atoms are kept from falling into the vdW potential. We have not reached a complete understanding of the surface modification mechanism yet. But we suspect that photoionized Cs-atoms may play a key role in the mechanism, since the effect is observed when atoms are irradiated by the violet laser under the MOT condition, where photoionization can occur efficiently through a two-step resonant ionization process [19].

The observed antibunching of fluorescence photons in figure 3(a) confirms that single atoms are detected using an optical nanofibre, and that the dwell time of single atoms in the observation region is longer than spontaneous emission lifetime. The observed decay in the correlation at much longer timescale as shown in figure 3(b) can be attributed to the dwell time of the single atom in the observation volume [15, 20]. The dwell time of 1.8 $\mu$s corresponds to an atom transit-length of about 200 nm, assuming an atom temperature of 100 $\mu$K. One may conclude that the fluorescence photons might be emitted by free atoms just passing through the vicinity of the nanofibre, since the estimated transit-length is quite a reasonable value for a free atom passing through a nanofibre with a diameter of 400 nm [9].

However, the peculiar feature observed for spectral characteristics for single atoms in figures 4(a)–(c) cannot be understood straightforwardly. Such a sharp dip, narrower than the natural width, may suggest the existence of some kind of quantum interference. As far as the authors know, a free two-level atom probed by using just one laser beam cannot account for such a sharp spectral dip. Similar behaviour might be expected for a vacuum Rabi-splitting spectrum, but in the present system we do not have any cavity, and this is not a reasonable explanation. Similar behaviours are expected for the spectrum with electromagnetically induced transparency (EIT) [21], but we do not have any coupling laser beam to realize the EIT condition.

One possible scheme to explain the observed spectral characteristics, may be a V-type three-level system discussed in [22], which is beyond the free two-level atom picture. The scheme consists of two upper levels, closely spaced within the radiative broadening, and one lower ground level, and the upper levels spontaneously decay to the lower level with the same rate leading to a destructive interference at the centre. For this scheme, we calculate the excitation spectrum by solving the density matrix equations under stationary conditions. Calculated spectra for three Rabi frequencies are overlaid on the observed ones in figures 4(a)–(c) with dashed lines. Spacing between the upper levels is assumed to be 1.5 MHz. The sharp dips are reproduced, and the relative spectral intensities are also well reproduced. Regarding the Rabi frequencies, we have used 1.4 times smaller values than those for the $F = 4 \leftrightarrow 5$ two-level transition to keep the total decay rate of the upper state equal to the value for the $F = 5$ level. This smaller Rabi frequency also corresponds to the Rabi frequency assumed to explain the observations in figure 3(a).

The above V-scheme understanding suggests that the atom might be trapped in a tiny potential in the close vicinity of the nanofibre surface, and, moreover, that the atomic motion is quantized, resulting in the two motional sub-levels for the excited electronic state so that the
V-type three-level scheme can be realized. Naturally, two sub-levels might also be created for the ground electronic state, which may produce two closely lying V-schemes. Note that the depths for the central dip can be better reproduced by two closely lying V-schemes. However, one obvious question is how such a tiny potential can be created in the close vicinity of the nanofibre surface. We suspect that the mechanism may be related to the surface modification by the violet laser irradiation, but we have not reached any definitive mechanism yet. We should mention that the vdW potential does not account for such trapping potential. The vdW potential would result in a much more complicated spectrum [18], due to the existence of many vibrational levels. For such a situation, any quantum interference, which may induce a sub-natural sharp dip, will be washed out.

5. Conclusion

We have shown that single atoms can be detected using an optical nanofibre, and that single photons spontaneously emitted from the atoms can be readily guided into a single-mode optical fibre. We have found that single atoms around the nanofibre reveal a very prominent spectral feature, that is, the excitation spectrum splits into two peaks with a very small separation smaller than the natural linewidth. The observed spectrum can be explained more naturally by assuming a trapped atom model than assuming a free two-level atom. However, regarding the mechanism to fully explain the observations, we have not reached any definitive conclusion yet. In order to clarify the details of the mechanism, further experimental and theoretical work is needed. Although the observations have not been fully understood yet, we anticipate that the present results may trigger research to bridge single-atom physics and optical-fibre technologies, and may give a new perspective for quantum-information technologies. Also the present finding may be extended to various surfaces and various atomic and molecular species.

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