OBSERVATION OF TWO-PHOTON EXCITATION FOR THREE-LEVEL ATOMS IN A SQUEEZED VACUUM

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Abstract

The two-photon transition ($6S_{1/2}{\rightarrow}6D_{5/2}$) of atomic Cesium is investigated for excitation with squeezed vacuum generated via nondegenerate parametric down conversion. The two-photon excitation rate (R) is observed to have a non-quadratic dependence of $R = aI^2 + bI$ on the incident photon flux (I), reflecting the nonclassical correlations of the squeezed vacuum field.

1 Introduction

Over the last two decades, there has been great progress in the generation and application of manifestly quantum or nonclassical states of the electromagnetic field. Spectroscopy with such nonclassical light can reveal new optical phenomena associated with the interaction between the nonclassical fields and matter. In this paper, we report the first experimental observation of such a novel field-matter interactions, namely two-photon atomic excitation using squeezed vacuum light.

It is well known that the two-photon excitation rate (R) can often be expressed in terms of the second-order correlation function of the driving field [1]. For classical light, this rate depends quadratically on the incident photon flux (I). In contrast, it is theoretically predicted that the quantum correlations of a squeezed state can enhance this rate so that it depends linearly on I in the limit of small photon flux [2, 3, 4, 5]. More generally, the two-photon excitation rate versus incident photon flux of a squeezed vacuum field is well approximated by the combination of quadratic and linear components, as $R = aI^2 + bI$. As a realization of this theoretical prediction, we have investigated the two-photon transition ($6S_{1/2}{\rightarrow}6P_{3/2}{\rightarrow}6D_{5/2}$) for trapped atomic Cesium with squeezed vacuum light, and found a non-quadratic dependence of the excitation rate on the incident photon flux.

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2 Experiment

The squeezed vacuum light is generated from a tunable optical parametric oscillator (OPO) [6] pumped under subthreshold condition. The pump beam is the second harmonic of a Ti:Sapphire laser (λ=883 nm), the frequency of which is locked ($\pm$0.3 MHz) to the two-photon resonance $6S_{1/2}$,F=4 $\rightarrow$ $6D_{5/2}$,F=6 of atomic Cesium. The OPO is tuned to generate two frequencies (λ_1=852 nm and λ_2=917 nm) in resonance with the transitions $6S_{1/2}$,F=4 $\rightarrow$ $6P_{3/2}$,F=5 and $6P_{3/2}$,F=5 $\rightarrow$ $6D_{5/2}$,F=6, respectively. The doubly resonant condition of the OPO cavity (linewidth $\sim$8 MHz) to the two frequencies is identified by monitoring the parametric gain of an auxiliary beam from a diode laser at 852 nm which is locked ($\pm$0.3 MHz) to the $6S_{1/2} \rightarrow 6P_{3/2}$ resonance.

The output from the OPO is focused with a waist of $\sim$10 μm onto Cesium atoms in a magnet-optic trap (MOT) [7], which has a diameter of $\sim$200 μm. The population of the upper excited state ($6D_{5/2}$) is measured by observing the fluorescence at 917 nm ($6D_{5/2} \rightarrow 6P_{3/2}$) with an avalanche photodiode. By chopping the trapping beams of the MOT at 4 kHz, we measure two counting rates R_1 and R_2, the rates with the trapping beams on and off, respectively. Since the trapping beams provide appreciable population of $6P_{3/2}$, R_1 provides a measure of the incident photon flux at 917 nm, while R_2 is proportional to the two-photon excitation rate driven by the squeezed vacuum field at 852 nm and 917 nm.

Since the counting rate R_2 is very small ($\leq 1\ s^{-1}$) in the region of interest, special care has been taken to eliminate and to determine accurately residual backgrounds. We used two different techniques to measure the background for a particular run. First, the magnetic field for the MOT is switched off thus eliminating the trap. Second, an interference filter is placed to block the 852 nm beam thus eliminating the two-photon transition. In both cases, no difference in results is discerned within an accuracy of $\pm$0.1/s, indicating that there are no systematic offsets in the background levels within the precision of our data.

3 Results and Discussion

We have performed several individual runs of the experiment, each of which took up to 10 hours for the actual data acquisition. In Fig. 1 is shown one example of the experimental plot of R_2 vs. R_1, where (a) and (b) are taken with approximate coherent state excitation and squeezed vacuum excitation, respectively [8]. For the coherent state excitation (a), the dependence of R_2 on R_1 is well described by the simple quadratic relation, $R_2 = a'R_1^2$, with the significance level (α) of 0.86. However, for the squeezed vacuum excitation (b), the data tend to depart from the quadratic form in the low intensity region. In fact, the data for (b) are well described by a combination of quadratic and linear components, $R_2 = aR_1^2 + bR_1$, with the significance level of α=0.69, while the simple quadratic fit can be rejected because of the far smaller value of the significance level (α=0.07). In Table I, significance levels calculated for five recent experimental runs are summarized. One can see that the function $R_2 = aR_1^2 + bR_1$ produces the largest significance levels for every experimental run and that it is the only acceptable one. The existence of the linear component is consistent with the theoretical predictions [2, 3, 4, 5], which take account of the quantum correlations between the two fields (λ_1 and λ_2) of the squeezed vacuum.

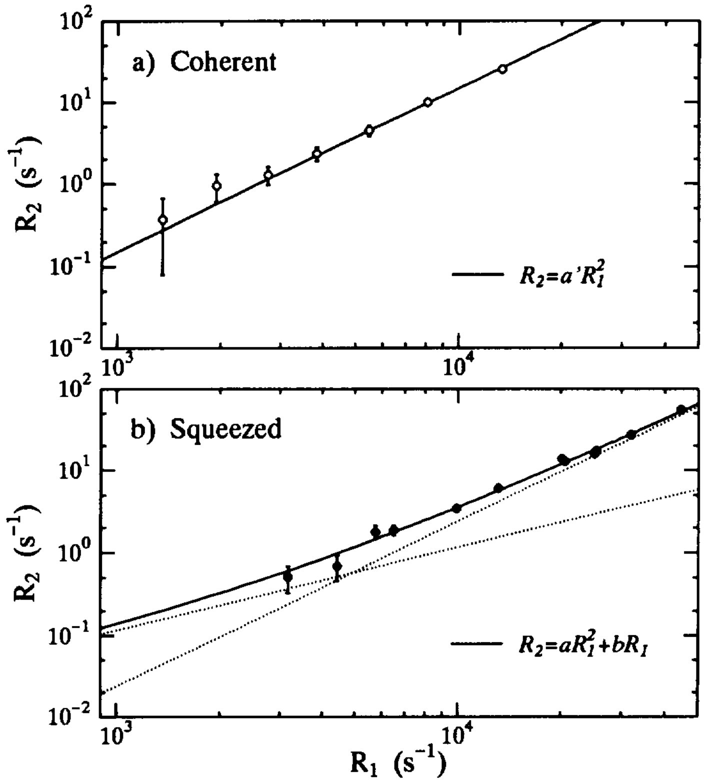

FIG. 1. Two photon excitation rate (R_2) versus excitation intensity (R_1). (a) Excitation with approximately coherent light, and (b) excitation with squeezed vacuum. Solid curves indicate the fitted functions of $R_2 = a'R_1^2$ and $R_2 = aR_1^2 + bR_1$ for (a) and (b), respectively. Dotted curves for (b) are asymptotic linear and quadratic components.

In addition to the measurement of R_1 and R_2, we also record the parametric gain (G) of the OPO at 852 nm. By using the relationship between G and R_1 (or R_2), one can deduct the "knee" position where the linear and quadratic components give equal contributions [9]. The average value of the knee position G_{knee} for five experimental runs is G_{knee}=1.36±0.09, and each value shows reasonable consistency within the statistical error. This value is to be compared with the theoretical expectation G_{knee}=1.7, which is obtained from numerical integration of the Master Equation appropriate to our system [10]. Although the measurements give somewhat smaller values and the reason for that is not clear at present [11], the agreement between the measured and theoretical values of the knee position is not unreasonable. Furthermore, the consistency of the measured values strongly indicates that the observed dependence of R_2 on R_1 is due to the properties of the light emerging from the OPO, and not to some spurious effects.

TABLE I. Significance levels for three trial functions $R_2 = aR_1^2 + bR_1$, $R_2 = a'R_1^2$, and $R_2 = a''R_1^2 + c$. (A) to (E) are the values for particular experimental runs, and (Total) for all the data scaled together as described in the text.

Experiment	$R_2 = aR_1^2 + bR_1$	$R_2 = a'R_1^2$	$R_2 = a''R_1^2 + c$
A	0.001	0.0002	0.002
B	0.69	0.07	0.44
C	0.33	0.0005	0.05
D	0.89	0.32	0.48
E	0.51	0.004	0.11
Total	0.03	2×10^{-10}	0.003

By using the simultaneous measurements of R_1, R_2, and G, one can combine all our experimental data onto a common scale, so that the measured variables (R_1, R_2, and G) fit the theoretical value by means of a least-squares minimization. As shown in Table I (Total), the experimental data thus scaled together can be fit by the function $R_2 = aR_1^2 + bR_1$, with the largest value for the significance level. Meanwhile, the fit with the functions of simple quadratic ($R_2 = a'R_1^2$) and quadratic plus constant ($R_2 = a''R_1^2 + c$) should be rejected because the significance levels for such fits are much smaller. Thus, we conclude that the experimental data do exhibit the predicted linear component of the two-photon excitation rate versus incident photon flux. We believe that the linear dependence is characteristic of the nonclassical nature of the squeezed vacuum excitation, because we can exclude the possibility of a linear dependence for classical fields in several broad cases [12].

In conclusion, we have made the first observation of a nonclassical effect on atomic excitation with a squeezed vacuum field. Our observations reveal a new regime of the field-matter interaction where the nonclassical nature of the field plays a role not heretofore realized.

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References

[1] B. R. Mollow, Phys. Rev. **175**, 1555 (1968).

[2] J. Janszky and Y. Yushin, Phys. Rev. A **36**, 1288 (1987).

[3] J. Gea-Banacloche, Phys. Rev. Lett. **62**, 1603 (1990).

[4] J. Javanainen and P. L. Gould, Phys. Rev. Lett. **41**, 6247 (1991).

[5] Z. Ficek and P. D. Drummond, Phys. Rev. Lett. **43**, 6247 (1992).

[6] E. S. Polzik, J. Carri, and H. J. Kimble, Phys. Rev. Lett. **68**, 3020 (1992), Appl Phys. B **55**, 279 (1992).

[7] N. Ph. Georgiades, E. S. Polzik, and H. J. Kimble, Opt. Lett. **19**, 1474 (1994).

[8] Absolute values of the excitation rate for the squeezed vacuum and those for the coherent light should not be directly compared because of experimental difficulties in reproducing operating conditions from one run to the next.

[9] Another equivalent definition of the "knee" is the position where the slope $d(\log R_2)/d(\log R_1)$ is 1.5.

[10] A. S. Parkins, unpublished (1993-1995).

[11] Small OPO detuning is one suspected reason.

[12] N. Ph. Georgiades, E. S. Polzik, K. Edamatsu, and H. J. Kimble, to be published.