Nonclassical Excitation in Spectroscopy with Squeezed Light

An experiment with squeezed light has demonstrated a new type of nonclassical effect: Correlated two-photon absorption can produce a two-photon excited population with a linear intensity dependence.

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The amplitude and phase of all light fields are subject to fluctuations. Much of this irregularity arises from random and uncontrolled changes that occur in any light source and lead to random changes in the light wave's frequency and amplitude. Most of these factors can in principle be eliminated by a careful design of the light source. Even if all of these defects in the source are removed, however, light fields are still subject to fluctuations arising from the laws of quantum mechanics. Even in a highly stabilized laser, the resulting coherent electromagnetic field has an uncertainty in the phase and magnitude of its amplitude. All conventional sources of light fields have at least this noise level, which, in a fully quantum description, arises from vacuum fluctuations. At one time it was believed that this vacuum or coherent-state noise could not be eliminated.

About 15 years ago the concept of "squeezed states" was introduced into radiation theory, demonstrating how the uncertainty in the field amplitude could be reduced below the quantum noise level by a "Heisenberg trade-off" between complementary operators. For example, the uncertainty of the phase could be decreased at the expense of increased fluctuations of the magnitude. This possibility for obtaining light fields whose fluctuations are less than those expected from stabilized lasers has attracted a great deal of attention.[1] (See PHYSICS TODAY, August, page 18, for a brief account of quantum state reconstruction of squeezed light, and June, page 18, for research on squeezed phonon states.) Squeezed states of light require field quantization for their explanation; they cannot be understood by using semiclassical techniques that assume a classical electromagnetic field interacting with quantized atoms or detectors. They were first studied by theorists interested in their properties as generalized minimum-uncertainty states of two-photon coherent states.[2]

Figure 1 shows Wigner functions of three types of state, a thermal state (a), a coherent state (b) and a squeezed state (c). Notice that the horizontal spread of the coherent state is comparable to that of the thermal state. In contrast, the squeezed state is very narrow in one direction and wider in the other, indicating the Heisenberg trade-off in its uncertainties.

The first experimental realization of squeezed light was reported in the mid-1980s by Richart Slusher and coworkers[3] at AT&T Bell Laboratories. They used four-wave mixing of light in optical cavities crossing a beam of sodium atoms, and they observed a reduction of optical noise to about 10% below the vacuum level. Robert Shelby and coworkers[4] at IBM's Almaden Research Center used four-wave mixing in an optical fiber and generated squeezed light with a 12.5% reduction below the vacuum level.

Jeffrey Kimble and coworkers[5] at the University of Texas at Austin developed a frequency tunable source of a squeezed vacuum field with about 70% noise reduction below the vacuum level. In their experiment, a squeezed field is generated by means of nondegenerate parametric down conversion. In this nonlinear process, a pump mode of frequency $2\omega_0$ generates two modes, called the signal and idler modes, of frequencies ω_1 and ω_2 such that $\omega_1 + \omega_2 = 2\omega_0$. In experimental terms, squeezed vacuum is the squeezed state of light generated by an optical parametric oscillator operating below threshold—that is, below the level at which a field of significant intensity is produced. For such a state, one can measure the instantaneous electric field by using techniques such as homodyne detection. (See PHYSICS TODAY, August, page 18.)

The experimental successes in generating squeezed light make possible its use in various applications. The earliest applications to be considered were the reduction of quantum noise in optical communications and the detection of gravitational waves by optical interferometry.[1] Several other applications of squeezed light have been proposed, including computing, cryptography, spectroscopy, laser technology and high-resolution measurement. Kimble's group at Caltech demonstrated high-resolution measurements using squeezed light[6] in 1992. They reported an improvement in sensitivity of 3 dB beyond the usual quantum limit in the Doppler-free detection of hyperfine transitions in cesium.

Nonclassical correlations

The photons produced in squeezed light experiments have strong nonclassical correlations. As discussed in more detail in box 1 on page 36, these correlations are characterized by dimensionless parameters $N(\omega)$ and $M(\omega)$. For any selected mode of the light, one can define quadrature

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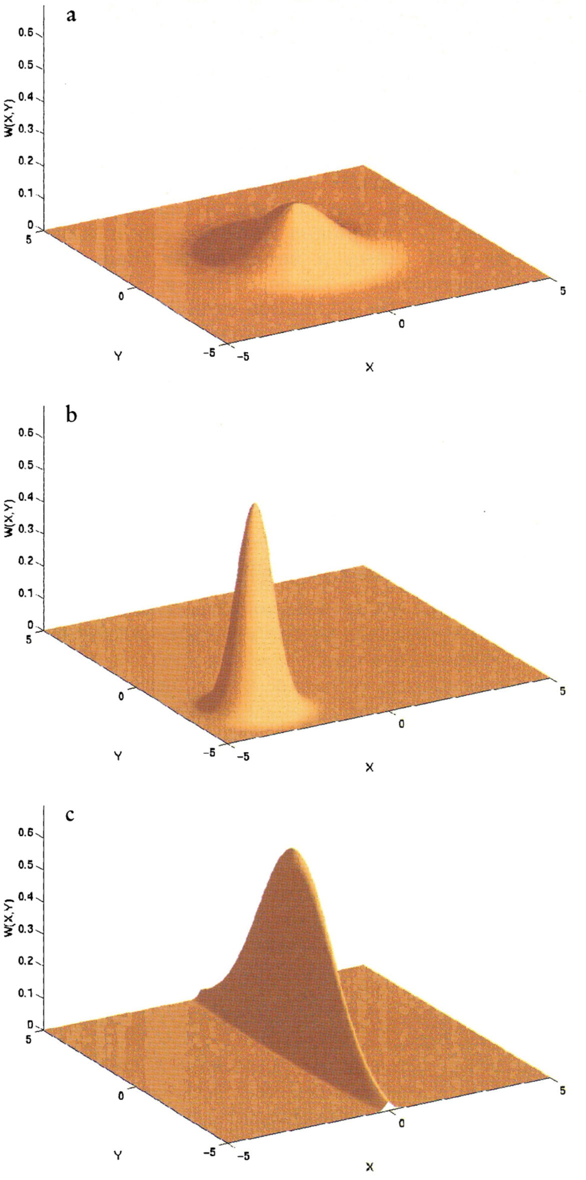

FIGURE 1. WIGNER DISTRIBUTION FUNCTIONS, $W(X,Y)$, for a thermal field (**a**), a coherent field as typically produced by a standard laser (**b**) and a squeezed field (**c**). The squeezed nature of the state in **c** is apparent from its narrowness in the X quadrature. The quadratures X and Y are analogous to position and momentum for a mechanical oscillator. The Wigner distribution function is a generalization of the wavefunction, suitable for describing mixed quantum states as well as pure ones.

operators X and Y, analogous to the $\hat{x}$ and $\hat{p}$ operators of a quantum particle in one dimension. The uncertainty relationship for these quadrature operators is

$$\Delta X \, \Delta Y \geq \frac{1}{4}. \qquad (1)$$

In figure 1 the horizontal axes correspond to these two operators. The squeezed state shown in the figure corresponds to a state that is maximally squeezed in the X quadrature—$M = \sqrt{N(N+1)}$—and that has large photon numbers, for which $N \gg 1$. Maximally squeezed states saturate the Heisenberg limit (that is, $\Delta X \, \Delta Y = 1/4$) and for large N, $\Delta X \approx 1/4\sqrt{N}$. In other words, these states have the minimal uncertainty product, and at large photon fluxes the uncertainty of the squeezed quadrature is much smaller than the vacuum level.

Although this reduction of uncertainty is the usual signature of a squeezed state, it is not the only signature. In addition to this effect, the two-photon coherence term $|M|$ is increased. In the output of a standard laser, $|M| = N$ and the normalized two-photon correlation function $C(\omega) \leq 1$. For a maximally squeezed field, however,

$$C(\omega) = \frac{1 + N(\omega)}{N(\omega)}. \qquad (2)$$

The result is a highly nonclassical correlation as graphed in figure 2, which is greatest for $N < 1$. Thus, unlike the usual squeezing measurements, there is a maximal nonclassical effect at *small* photon flux levels (that is, less than one photon per second in each one-Hertz interval of frequency).

In summary, the presence of squeezing leads to both an anisotropic distribution of the noise in the phase space, with the noise significantly reduced in some directions, and a relative increase in two-photon correlations. The nonclassical character of the correlations is manifested by the term 1 in equation 2, which arises from the quantum nature of the field, so that the operators $a^\dagger(\omega_i)$ and $a(\omega_i)$ do not commute. For a classical field, the correlations can have the maximum value $|M(\omega_i)| = N(\omega_i)$. Therefore, any effect arising from the excess correlations over that for a classical field has a nonclassical character.

Squeezed light spectroscopy

The idea of nonclassical atomic spectroscopy, or squeezed light spectroscopy, was originally due to Crispin Gardiner,[7] who showed theoretically that nonclassical effects, such as inhibition of the decay of the atomic dipole moment (which is sensitive to photon correlations), could occur in spontaneous emission from two-level atoms. However, no changes in atomic populations were predicted, making the nonclassical effects in two-level atoms difficult to observe experimentally.

Two-photon absorption between atomic levels of the same parity has attracted considerable interest in atomic spectroscopy. In general, a two-photon absorption in a system of atoms irradiated by an external field can occur as a two-step process with one photon absorbed in each step. Since the intensity of the field is proportional to the number of photons, a two-photon process normally exhibits a quadratic dependence on the intensity of the exciting field. The close connection between squeezed light and two-photon processes[2] suggests that two-photon transitions in multilevel atoms may be especially sensitive to the correlations characteristic of squeezed light. In the case of three-level atoms, work in this area was pioneered by Julio Gea-Banacloche[8] and also by Juha Javanainen and Philip Gould,[9] who showed that the two-photon transition rate could depend linearly on intensity (instead of quadratically) in a transient regime. But could this lead to a nonclassical steady state population that also depends linearly on intensity?

Let us consider a three-level atom in a cascade configuration (see figure 3) interacting with two coherent or thermal radiation fields of equal intensities I, each coupled

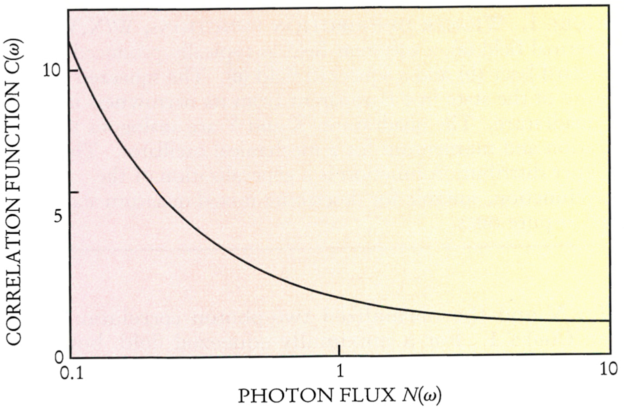

Figure 2. Nonclassical behavior develops at very low levels of photon flux $N(\omega)$, because of the dramatic increase in the relative correlation $C(\omega)$, which can be at most 1 for a field admitting a classical description.

to a one-photon transition. In this case the two-photon transition $|0\rangle \to |2\rangle$ appears as a two-step transition $|0\rangle \to |1\rangle \to |2\rangle$. At low intensity, each one-photon transition rate is proportional to intensity I, resulting in a quadratic intensity dependence for the two-photon transition rate. In the steady state, the population ρ in level $|2\rangle$ is therefore proportional to I^2.

Recent theoretical[10,11] work has showed that correlated photon pairs, characteristic of squeezed light, can indeed affect the two-photon transition rate in a nonclassical way that causes a linear rather than quadratic intensity dependence of the population ρ. Detailed calculations show that the population exhibits both linear and quadratic dependence on the intensity when the atom is excited by a squeezed field. (See equation 14 in box 2 on page 37.) This dependence is in contrast to the purely quadratic intensity dependence produced by classical light sources.

For low intensities ($N \ll 1$), the linear term of equation 14 dominates, which is the direct manifestation of the modification of the two-photon absorption $|0\rangle \to |2\rangle$. In

Box 1. Correlations and Uncertainties of Squeezed Light

In experiments that produce squeezed light, the output modes exhibit strong nonclassical correlations of the emitted photons. These are characterized by correlation functions for the field-mode operators of the type

$$\begin{aligned} \langle a(\omega_i)\, a^\dagger(\omega_j)\rangle &= [N(\omega_i) + 1]\,\delta(\omega_i - \omega_j)\,, \\ \langle a^\dagger(\omega_i)\, a(\omega_j)\rangle &= N(\omega_i)\,\delta(\omega_i - \omega_j)\,, \\ \langle a(\omega_i)\, a(\omega_j)\rangle &= M(\omega_i)\,\delta(2\omega_0 - \omega_i - \omega_j)\,, \end{aligned} \tag{3}$$

where $a^\dagger(\omega)$ and $a(\omega)$ are the usual creation and annihilation operators (respectively) of a photon of frequency ω. Here, $N(\omega)$ has units of photons/[Hz·s], and so is a dimensionless quantity, giving the output photon flux per unit frequency in an external transverse mode that is matched to the internal cavity mode of the squeezing generator.

The parameters $N(\omega_i)$ and $M(\omega_i)$ describe the spectral intensity of the modes in photon units. When the output intensity is symmetric (relative to ω_0), the correlations between the output modes obey the inequality

$$|M(\omega_i)| \le \sqrt{N(\omega_i)[N(2\omega_0 - \omega_i) + 1]}\,. \tag{4}$$

It is customary to simplify the quantum analysis to a single longitudinal mode of an idealized cavity with frequency $2\omega_0$ in the external field. Suppose this gedanken cavity has single-mode electromagnetic annihilation and creation operators a, $a^\dagger$. We can define complementary second-quantized quadrature operators, X and Y, analogous to the $\hat{x}$ and $\hat{p}$ operators of the Heisenberg uncertainty relation. These are defined as

$$\begin{aligned} X &= \frac{1}{2}(a^\dagger + a)\,, \\ Y &= \frac{1}{2i}(a^\dagger - a)\,. \end{aligned} \tag{5}$$

They have commutation properties of

$$[X, Y] = \frac{1}{2i}\,. \tag{6}$$

This implies an uncertainty relationship

$$\Delta X\, \Delta Y \ge \frac{1}{4}\,, \tag{7}$$

where ΔX is the quadrature standard deviation. (The factor $\hbar$ is absorbed in the definition of the operators a and $a^\dagger$.)

These commutation relations are fundamentally due to the commutator between the electric and magnetic fields in the vacuum, expressed in a modal expansion form.

In a squeezed state of the type we are discussing here, the values of the standard deviations are given by

$$\begin{aligned} \Delta X &= \frac{1}{2}\sqrt{1 + 2(N - M)}\,, \\ \Delta Y &= \frac{1}{2}\sqrt{1 + 2(N + M)}\,. \end{aligned} \tag{8}$$

Here we assume that the quadrature maximum and minimum fluctuations are aligned with the X and Y axes, respectively. For a maximally squeezed state, $|M| = \sqrt{N(N+1)}$ and

$$\Delta X\, \Delta Y = \frac{1}{4}\,. \tag{9}$$

For a maximally squeezed state, squeezed in X and with large N, $\Delta X \approx 1/4\sqrt{N}$. In other words, these states have the minimal uncertainty product, but the uncertainty of any given quadrature can be much smaller than the vacuum level, at large N values.

In addition to this reduction of uncertainty, there is an increase in the two-photon coherence term, $|M|$. In the output of a standard laser (coherent but unsqueezed), the normally ordered correlations factorize, so that $|M| = N$. By comparison, consider the normalized two-photon correlation functions defined by

$$C(\omega) = \frac{|\langle a(\omega)\, a(2\omega_0 - \omega)\rangle|^2}{N(\omega)\, N(2\omega_0 - \omega)}\,. \tag{10}$$

In our idealized case of a maximally squeezed field, this ratio increases dramatically at low intensity. The result is a highly nonclassical correlation, which is greatest for $N < 1$, as can be seen from the following equation, which is applicable to our maximally squeezed gedanken mode:

$$C(\omega) = \frac{1 + N(\omega)}{N(\omega)}\,. \tag{11}$$

This is graphed in figure 2. Thus, there is a maximal nonclassical effect at photon flux levels of less than 1 $s^{-1}Hz^{-1}$.

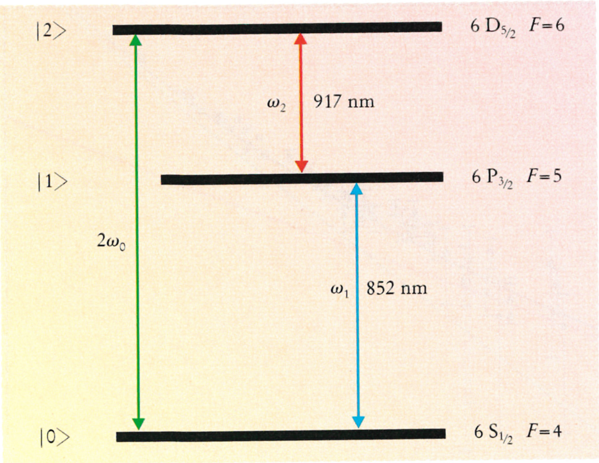

FIGURE 3. THREE-LEVEL ATOM in the cascade configuration. A two-photon transition between states $|0\rangle$ and $|2\rangle$ can occur in a single step when the fields have suitable correlations. The population ρ in state $|2\rangle$ can be affected by this process. The indicated spectroscopic states and frequencies are those relevant to the experiment that tested these effects in cesium atoms.

a squeezed vacuum field, the two-photon correlations enable the transition $|0\rangle \to |2\rangle$ to occur effectively in a "single step" proportional to N. Another way to think of this is that amplitudes add coherently (because of the term 1 in equations 2 and 4) for the $|0\rangle \to |1\rangle$ and $|1\rangle \to |2\rangle$ transitions, rather than incoherently.

This single-step two-photon process has an immediate effect on the populations of the atomic levels. For example, in this process, the intermediate level is not populated. This effect results in one-photon, as well as two-photon, inversions between the atomic levels. The Boltzmann distribution of the populations of the atomic states is violated. All of these effects are essentially novel in the field of spectroscopy.[10,12]

For larger intensities ($N \geq 1$), the quadratic term, characteristic of classical-field excitation, dominates and masks the linear, nonclassical term. Thus, to observe the departure from the quadratic intensity dependence, it is important to apply a squeezed vacuum field with low intensity, since this relative change is due to the correlation $C(\omega)$, which is maximal when $N < 1$.

Another factor, important from the experimental side, is the matching of the input squeezed modes to the vacuum modes coupled to the atom. This matching is characterized by the parameter η, which is 1 for perfect

Box 2. Steady State Populations

Consider the three-level system (shown in figure 3), driven by a broadband squeezed vacuum field, in which the squeezing bandwidths near the transition frequencies ω_1 and ω_2 are greater than the relevant natural linewidths.[10] (The squeezing bandwidth is the range of frequencies over which the driving vacuum field exhibits squeezed fluctuations.)

The steady state population in the upper state $|2\rangle$ is

$$\rho = \rho_{22} = \frac{\eta^2 N^2 W - \eta^2 |M|^2 (W - 1 - \alpha)}{W(3\eta^2 N^2 + 3\eta N + 1 - 3\eta^2 |M|^2)}, \tag{12}$$

where $\alpha = \Gamma_{21}/\Gamma_{10}$ is the ratio of the spontaneous emission rates Γ_{21} and Γ_{10} of the transitions $|2\rangle \to |1\rangle$ and $|1\rangle \to |0\rangle$, respectively, and $W = \alpha + \eta N(1 + \alpha)$, $N = N(\omega_1) = N(\omega_2)$ and $|M| = |M(\omega_1)| = |M(\omega_2)|$. The parameter η describes the matching of the incident squeezed vacuum modes to the vacuum modes coupled to the atom. For perfect matching, $\eta = 1$ whereas for imperfect matching, $\eta < 1$.

For a thermal field without correlations between photons, $|M| = 0$, and the populations have a thermal (Boltzmann-type) distribution, with

$$\rho = \frac{\eta^2 N^2}{3\eta^2 N^2 + 3\eta N + 1}. \tag{13}$$

For low intensities ($N \ll 1$), this is proportional to N^2, showing that in thermal fields the population ρ exhibits a quadratic dependence on intensity—as is also the case for coherent lasers.

For a minimum uncertainty squeezed vacuum field, for which $|M|^2 = N(N + 1)$, the population ρ is given by

$$\rho = \frac{\eta^2 N + \eta^2 N^2 (1 + \alpha)(1 - \eta)}{(\alpha + \alpha\eta N + \eta N)[3\eta N(1 - \eta) + 1]}. \tag{14}$$

This population exhibits both linear and quadratic dependence on the intensity of the squeezed vacuum field.

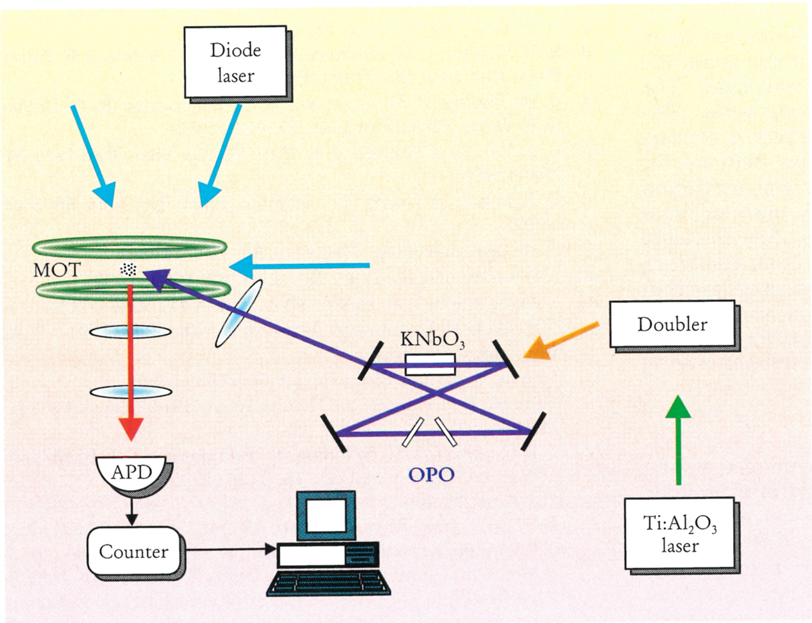

FIGURE 4. EXPERIMENTAL SETUP for observing the nonclassical dependence on intensity of the population ρ. The pump field (orange) of the frequency-doubled output of a titanium–sapphire (Ti:Al$_2$O$_3$) laser is transmitted through an optical parametric oscillator (OPO) cavity to produce a two-mode squeezed vacuum field (purple) of wavelengths 852 nm and 917 nm. This vacuum field is coupled to cesium atoms in a magneto-optic trap (MOT), whose optical part (blue) is produced by an 852 nm diode laser. The cesium's fluorescence (red) at 917 nm is recorded with an avalanche photodiode (APD) and analyzed by a computer.

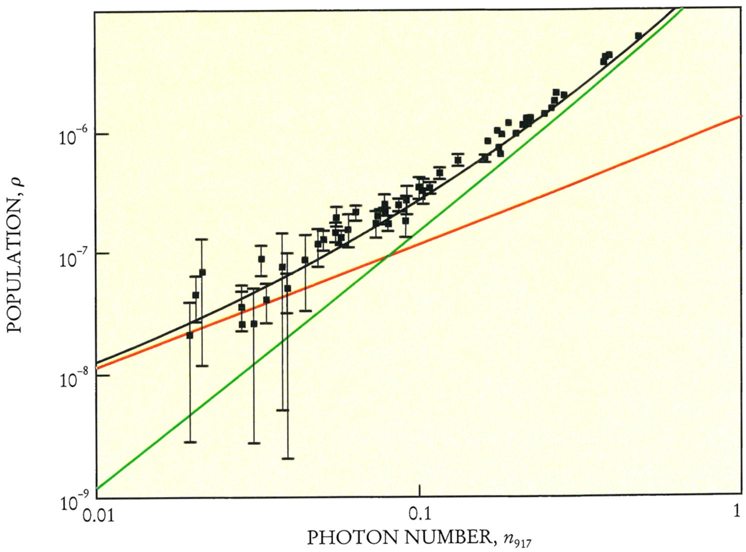

FIGURE 5. LINEAR AND QUADRATIC DEPENDENCE of the population ρ as a function of the photon number n_{917} at 917 nm. The black curve is obtained from equation 12, whereas the red and green lines indicate the linear and quadratic components, respectively. (From ref. 13.)

matching. However, as can be seen from equation 14, the linear dependence on the intensity is not destroyed by an imperfect matching ($\eta < 1$), but the population is reduced.

Experimental verification

In 1995 the linear dependence of the population ρ on the squeezing intensity N was observed experimentally by Kimble's group at Caltech,[13] in a difficult and pioneering experiment. In their experiment (see figure 4), the squeezed vacuum field was generated by an optical parametric amplifier whose output consists of two low-intensity but very strongly correlated beams of frequencies ω_1 and ω_2, symmetrically located about the carrier frequency $\omega_0 = (\omega_1 + \omega_2)/2$. The output exhibits nonclassical correlations between the two beams, as characterized by equation 2. This squeezed vacuum was then focused into a cloud of cesium atoms in a magneto-optic trap. The atomic cesium behaves as a three-level atom with transition wavelengths $\lambda_{21} = 917$ nm, $\lambda_{10} = 852$ nm. The two beams of frequencies ω_1 and ω_2 are tuned to these atomic transitions.

By monitoring the fluorescence at 917 nm (which is proportional to the population ρ), the experimental team observed that the population ρ deviated from the quadratic intensity dependence observed with a coherent light. The results are shown in figure 5 using a log–log scale. The observed departure from the slope of 2, which is characteristic for classical fields, gives compelling evidence for the quantum, or nonclassical, nature of the excitation process. As was predicted by the theory outlined here (and confirmed by computer calculations that included finite bandwidth effects[13]), the departure from the slope of 2 appears for low intensities of the squeezed vacuum field. For $N \gg 0.3$ the slope is more or less the same as the classical limit of 2. This somewhat smaller intensity for the change in slope is due to mode matching, and is also predicted by the detailed theory.

Conclusions

The effort to demonstrate the linear dependence on the intensity of two-photon transitions is part of a broader and very active field involving nonclassical excitation of atomic systems. Many interesting modifications of radiative properties of atoms in the presence of squeezed light have been predicted. Examples include inhibition of the atomic decay process, squeezing-induced transparency, population trapping, dispersive and subnatural profiles in the fluorescence spectrum and amplification without population inversion.[14] (See the article by Stephen Harris in PHYSICS TODAY, July, page 36, for discussion of some of these processes, using classical fields.) It should be noted that some of these features, unlike the linear dependence of the two-photon transition rate, are not limited to low intensities of the squeezed vacuum and appear even for very large intensities. Kimble's pioneering experiment demonstrates that nonclassical spectroscopy offers new physical effects not obtained with conventional radiation sources. Although this experiment was restricted to low intensities, it is a clear indication that there is more to nonlinear spectroscopy than meets the eye, when squeezed fields are involved.

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