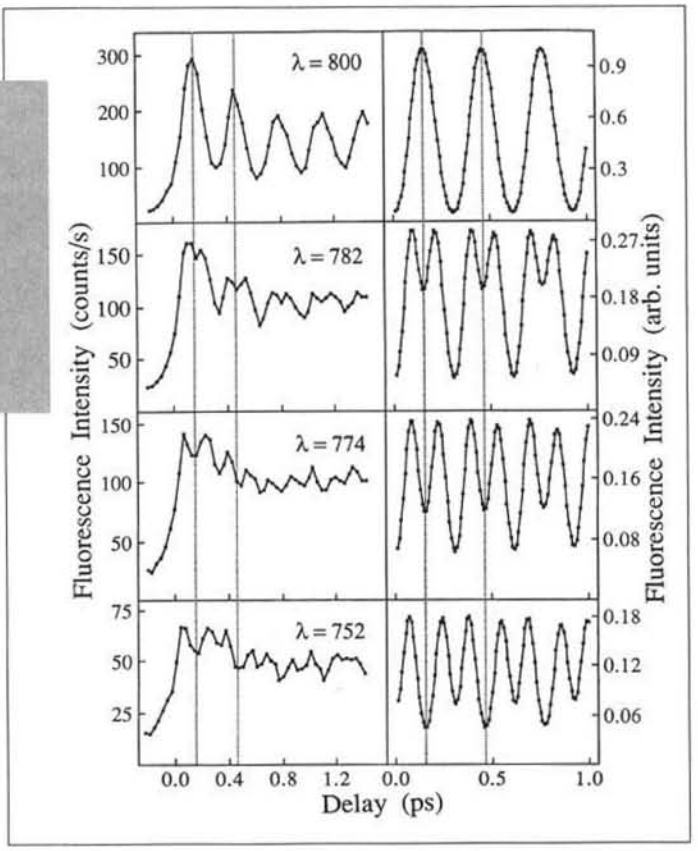

The experimental and theoretical time- and frequency-resolved spectrum of spontaneous emission. The motion of the mean position of the wave packet is clear from the changing temporal structure at different detection wavelengths (shown in nanometers) as discussed in the text.

electronic level, places the nuclei in a new electronic potential that has a different equilibrium nuclear separation. The nuclei are displaced in the new potential and begin to oscillate. At any time, the molecule may emit a photon and return to its ground electronic state. The wavelength of the emitted photon depends on the internuclear separation at the time of emission.

The results of our measurements are shown in the figure at right together with data calculated using the theoretical model. The emission at 800 nm occurs when the wave packet is at the classical outer turning point. The first maximum occurs 155 fsec after excitation, indicating that the nuclei take exactly one-half period to reach the outer turning point after being generated near the inner turning point, exactly the behavior of a classical vibration. As the spectrometer is tuned to a slightly shorter wavelength at 782 nm, a "splitting" of the beats is observed compared with the signal at 800 nm. Away from the turning points, double beats are observed since the wave packet passes any internuclear separation twice in each oscillation. The motion of the mean position is clear from the changing temporal structure at different detection wavelengths.

The data also illustrate the expansion and contraction of the wave packet that characterize a vibrational quadrature squeezed state.[3] The deep modulation of the quantum beats at 800 nm indicates that the wave packet is well localized. The expansion of the wave packet as it moves from the outer turning point towards the middle of the potential is evident from the decrease in the modulation and the reduction in the overall signal amplitude.

Our experiments show that time-resolved fluorescence spectroscopy is likely to be an important tool in studying the dynamics of wave packets in atoms, molecules, and semiconductors.

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Number-phase Uncertainty Relations

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The uncertainty principle lies at the heart of quantum mechanics. Any pair of noncommuting variables satisfy a form of uncertainty relationship, and this sets bounds on the measurement precision achievable for these quantities. In quantum optics, one such pair of variables are the photon number and the phase of an optical field. They satisfy an uncertainty relation $\Delta n \Delta\phi \geq (1/2)|\langle|\Psi[\hat{n}, \hat{\phi}]|\Psi\rangle|$ where $|\Psi\rangle$ represents the state of the field. One important point to note is that the lower bound of this relation depends on the state. Thus, one needs a way of determining the values of both the uncertainty product and the expectation value of the commutator $[\hat{n}, \hat{\phi}]$.

Two equivalent ways to describe the quantum properties of the phase of a field come from the Pegg-Barnett phase operator[1] and the Shapiro-Shepard probability operator measure.[2] A problem with using these phase formalisms is that there is currently no known way to directly measure the phase described by them. We have overcome this obstacle by inferring these phase distributions from measurements of the quantum mechanical state of the system.

In our experiments we use a balanced homodyne detector that measures the quadrature amplitude of a signal field.[3,4] We use a pulsed laser and measure the quadrature amplitude on each laser shot. By performing many such measurements, we build up probability distributions of the quadrature amplitude. By varying the phase, θ, of a local oscillator field, we change which field quadrature is measured. We are able to obtain probability distributions for many different quadratures, each parametrized by a different value of θ.

It was shown by Vogel and Risken that if one obtains probability distributions for quadrature amplitude on a continuous set of θ between 0 and π, it is possible to perform the inverse Radon transformation on these distributions and obtain the Wigner function of the field.[5] If one has distributions for a finite and discrete set of angles between 0 and π, one may use techniques developed in computer-aided tomography (CAT) to perform the inverse Radon transformation. Thus, using a CAT algorithm we are able to reconstruct the Wigner function of the field from our measured distributions. In quantum mechanics, the Wigner function contains all knowable information about the state of a system, so from it one can calculate distributions and/or moments of any quantity of interest. If the measured state is found to be pure, it is possible to construct the wavefunction of the field state, and we have done this for measured coherent-state fields.

In Figure 1(a) we show the Pegg-Barnett/Shapiro-Shepard phase distributions for coherent states of differing average photon number. As the photon number decreases, the phase becomes less well defined. In Figure 1(b), both the number-phase uncertainty product and the lower bound set by the commutator expectation are plotted. The uncertainty product is greater than the commutator expectation (as it must be since our method assumes the validity of quantum mechanics). The interesting point is that equality in the uncertainty relation is only achieved at very small and very large average photon number.

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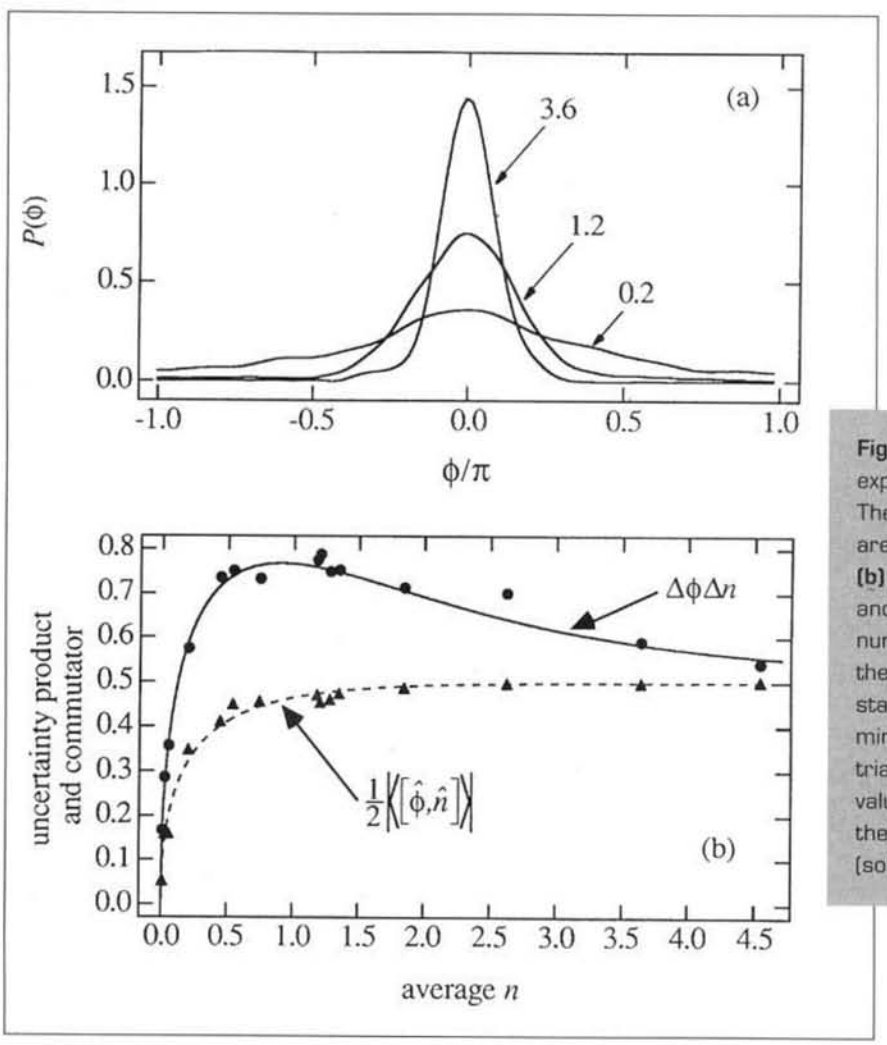

Figure 1. (a) The phase distributions for experimentally measured coherent states. The mean number of photons in these states are indicated.
(b) The number-phase uncertainty product and the expectation value of the number-phase commutator are plotted versus the mean number of photons in a coherent state. The circles are experimentally determined values for the uncertainty product; the triangles are experimentally determined values for the commutator. The curves are theoretical values for the uncertainty product (solid line) and the commutator (dashed line).