Interference between Optical Transitions

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We report observations of an interference between optical transition amplitudes for linear and nonlinear excitation of the mercury $6s\,^1S_0 \rightarrow 6p\,^1P_1$ transition. The transition probability is varied sinusoidally by changing the relative phase of the two fields inducing these distinct processes.

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In this Letter we report the observation of an interference between different optical processes which can individually lead to resonantly enhanced multiphoton ionization. This interference has been identified[1-3] as the cause of the suppression of multiphoton excitation of various atomic species when the vapor density is relatively high. Interfering processes were first considered theoretically by Manykin and Afanas'ev,[4] who calculated the two-photon excitation of a nonlinear medium used in two-photon resonantly enhanced third-harmonic generation. Suppression of multiphoton ionization[5] and multiphoton absorption[6] at high vapor densities were first observed in 1977. Since that time, the correlation between the suppression of multiphoton excitation or ionization and generation of harmonic waves in the medium[7-10] has been studied, as well as the competition between four-wave mixing and amplified spontaneous emission,[11] the suppression of multiphoton ionization by amplified spontaneous emission,[12] and the suppression of two-photon excitation in sum-frequency-mixing processes.[13]

In the present work we observe the interference between two ionization processes in mercury (see Fig. 1), each leading to the same continuum state and each resonantly enhanced by the $6p\,^1P_1$ intermediate state. The first process is a five-photon process using light at $\lambda = 554$ nm. This process is resonantly enhanced due to the proximity of 3 times the laser frequency to the $6s\,^1S_0 \rightarrow 6p\,^1P_1$ transition frequency. The second process is a three-photon process using one photon at $\lambda/3$ and two photons at λ. The single-photon step involving the ultraviolet photon is in near resonance with the same transition in mercury. Thus the interference which we observe is due to the choice the atom must make to participate in one interaction or the other. This interference is clearly revealed by measuring the dependence of the ionization probability of the atomic mercury on the relative phase between the two fields. For purposes of clarity, we discuss the excitation process to the $6p$ state rather than the ionization process directly. The arguments are easily extended to the latter process. Under conditions of concurrent excitation by the two fields, E^{uv} and E^{vis}, the net transition rate is expected to be given by

$$W_{1,3} = \frac{2\pi}{\hbar^2} |\mu E^{\mathrm{uv}} e^{i\phi_1} + \mu^{(3)} (E^{\mathrm{vis}} e^{i\phi_2})^3|^2 g(\Omega_{fg} - 3\omega) .$$

Here μ and $\mu^{(3)}$ are the transition moments for linear and three-photon processes, respectively, and $g(\Omega_{fg} - 3\omega)$ is a line-shape function. The phase, ϕ_i $(i = 1,2)$, of each field has been retained in this expression, indicating that there will be a term in the transition probability which varies as $\cos(3\phi_2 - \phi_1)$.

In the case of excitation by plane waves, the single-photon and three-photon transition amplitudes can be matched everywhere by the adjustment of the field amplitudes E^{uv} and E^{vis}, resulting in a maximum depth of modulation of 1. When using focused laser beams to photoionize the atoms, we must consider that the relative amplitudes of the two beams, as well as their relative phase, will vary the focal region. This will limit the depth of modulation of the interference fringes to a value of less than 1. If we assume lowest-order Gaussian modes, the transition rate at a distance z from the focus

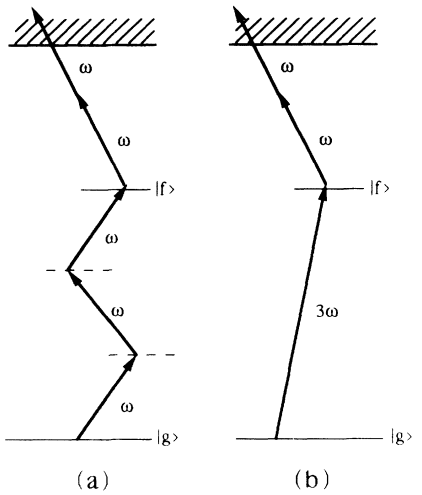

FIG. 1. The two processes which interfere in this observation. The transition $|g\rangle \rightarrow |f\rangle$ is three- and one-phonon allowed, as shown in (a) and (b).

is proportional to

$$W \propto \left| \frac{E^{uv}\mu e^{i\phi_1}}{[1+(z/z_0)^2]^{1/2}} \exp\left(-\frac{3r^2}{w^2}\right) \exp\left[-i\left(k^{uv}z+\tan^{-1}\frac{z}{z_0}\right)\right] + \frac{(E^{vis}e^{i\phi_2})^3\mu^{(3)}}{[1+(z/z_0)^2]^{3/2}} \left\{\exp\left(-\frac{r^2}{w^2}\right)\exp\left[-i\left(k^{vis}z+\tan^{-1}\frac{z}{z_0}\right)\right]\right\}^3 \right|^2 , \quad (1)$$

where $w(z)$ is the $1/e^2$-intensity beam radius, k is the propagation constant, and $z_0=\pi w(0)/\lambda$ is one-half the confocal parameter. Under our experimental conditions z_0 is the same for the two beams, while the beam radius for the uv beam is $1/\sqrt{3}$ that of the visible beam. Since the three-photon process depends on the cube of the visible field, the z dependence of the magnitude of this term of the transition amplitude varies as $[1+(z/z_0)^2]^{-3/2}$ while the phase varies as $\exp[-3i\tan^{-1}(z/z_0)]$. Therefore, the magnitude of the two processes can be matched at, at most, two locations symmetrically placed about the focus, and the phase of the two transition amplitudes varies through the focal region over a range of 2π. The interference can still be quite visible, however, because of the high-order intensity dependence of the process, confining ionization to the center of the focal region. This effect can be calculated by integrating the expression for the ionization rate [Eq. (1)] over the entire focal region. Since two additional photons must be absorbed to ionize the mercury, we must weight Eq. (1) by an additional factor of I^2, yielding an average ionization rate of

$$W \propto (1+\tfrac{5}{8}M^2)+\frac{M\cos(3\phi_2-\phi_1)}{2} ,$$

where $M=(E^{vis})^3\mu^{(3)}/E^{uv}\mu$ represents the relative contribution of the two processes at the beam waist on axis. When $M=1$, the magnitude of the transition amplitudes are matched at the center of the focus, but the linear process will be stronger at all other locations. For $M=2$, the magnitudes are matched at $z=\pm z_0$. Inside the focus the three-photon process is stronger, while for $|z|>z_0$, the linear process is stronger. The depth of modulation has a maximum value of $1/\sqrt{10}$ at $M=(\frac{8}{5})^{1/2}$, and is within 10% of this maximum value over the range from $M=\frac{3}{4}$ to $M=2$.

The experiment is carried out in a cell consisting of three chambers, shown in Fig. 2. Coherent ultraviolet radiation at 185 nm is generated in the first chamber, containing mercury at a relatively high pressure of $\sim$100 mtorr (cold-finger temperature=80 °C) by focusing in laser radiation at 554 nm using a 20-cm focal-length lens. The radiation produced by third-harmonic generation, resonantly enhanced by the nearby $6p$ state, has a well-determined phase with respect to the laser fundamental. The visible and ultraviolet beams are collimated and refocused into the third chamber using a pair of spherical mirrors with a uv-enhanced aluminum coating (focal length=25 cm). The third chamber also contains mercury vapor, but at a much lower pressure ($\sim$2–3 mtorr) than that of the first cell by virtue of the cold-finger temperature of $\sim$30 °C. The relatively low vapor density in this cell is important to minimize the amount of third-harmonic radiation generated there. The second cell is used to introduce a phase shift between the two laser beams. By varying the density of the argon gas in this cell, the phases of the visible and uv beams undergo a shift of magnitude $\phi_2=2\pi l\Delta\rho n^{554}/\lambda$ and $\phi_1=6\pi l\Delta\rho n^{185}/\lambda$, respectively, where l is the path length in the argon chamber, $\Delta\rho$ is the change in density in amagats, and n^{554} and n^{185} are the refractive indices of argon gas at STP conditions at 554 and 185 nm, respectively. The relative phase between the two transition amplitudes thus varies as

$$3\phi_2-\phi_1=\frac{6\pi l\Delta\rho}{\lambda}(n^{554}-n^{185}) . \quad (2)$$

Observation of the interference fringes depends critically on several important features of the experiment, such as spatial coherence of the laser and beam overlap in the focal region. The beam produced by the homemade Littman-style short-cavity laser is a nearly TEM_{00} Gaussian beam operating on two to three longitudinal modes separated by the free spectral range of the laser of $\sim$3.3 GHz. After two stages of amplification in longitudinally pumped Brewster-angle dye cells, the laser beam has a pulse energy of $\sim$4.5 mJ and a pulse duration of less than 15 nsec, and is somewhat elliptical in shape with beam diameters of 2.2 and 1.3 mm along the major and minor axes, respectively. A lens cannot be used to focus the two laser beams into the third chamber because of the chromatic aberration it would introduce. An astigmatism due to the off-axes reflection from the mirrors, if present, would not change the depth of modulation of the interference since the overlap of the wave fronts would not be affected, but would serve to decrease the total ionization signal. The degree of astigmatism is made negligible by keeping the angle of incidence of the laser less than 3°. The fused-silica windows between the two focal regions have wedge angles less than 3 arc sec, limiting the separation of the uv and visible beams at the second focus due to dispersion to less than 1 μm. The beam radius at the focus is calculated to be $\sim$20 μm. (Measurements of the transmission of the focused beam by a 50-μm-diam pinhole provide an upper bound of 40 μm. There appear to be shot-to-shot fluctuations of the beam position which limit our measurements of the size

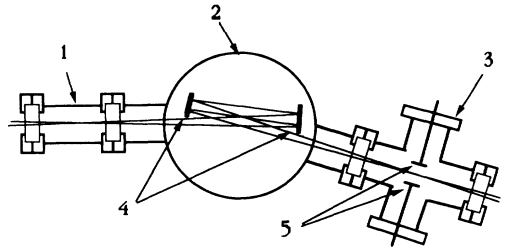

FIG. 2. The three-chamber cell. Chamber 1 contains mercury at a high density (~100 mtorr), 2 contains argon gas at a variable pressure (0–38 torr), and 3 contains mercury at a low density (~2–3 mtorr). Chamber 2 contains (4) a pair of uv-enhanced aluminum-coated spherical mirrors, and the ionization signal is measured using (5) a pair of biased platinum collection plates.

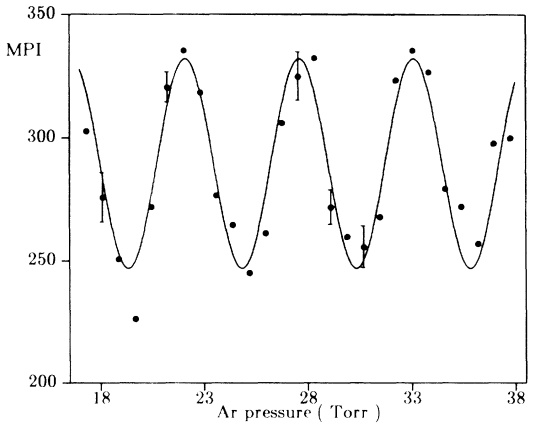

FIG. 3. Ionization signal measured as a function of argon pressure in chamber 2. Solid line indicates a best fit to the data. Error bars showing 1 standard deviation of the mean are shown for a few data points.

of the focused beam.) The two fields can also be displaced by the windows if the incidence angle is not sufficiently small. We calculate that a 1° angle of incidence will result in a 10-μm beam separation at the focus, so beam alignment must be maintained to less than this value for good fringe visibility. In practice, alignment must be much better than this estimate indicates, an observation which still needs to be explained.

The cell chambers containing the mercury were constructed of stainless steel, with O-ring seals for the windows. The vapor pressure of the mercury was maintained by controlling the temperature of a cold finger with the cell body held at a slightly higher temperature. Temperature fluctuations during a data run were typically $< \pm 0.3\,^{\circ}$C. Temperature stability in the first cell is important to maintain the magnitude and phase of the third-harmonic radiation produced there, while in the last cell stability is important since the signal strength is proportional to the mercury density.

The ionization signal is measured by collecting the electrons produced in the focal region by a pair of platinum parallel plates with a cross-sectional area of 1 cm × 1 cm. One plate is grounded to the cell body, and electrons are accelerated toward the other plate by a +24-V bias. The plate separation is 1 cm. The electron pulse is ac coupled into a fast (3-nsec rise time) transimpedance preamplifier with a gain of 25 mV/μA, and integrated by a gated integrating analog-to-digital converter. Data are accumulated by a PC AT laboratory computer. The laser power is also monitored by the detection system, and only data for laser powers within a $\pm 5\%$ range are accepted. The data are shown in Fig. 3. Each data point represents the average of 60–80 laser shots. The error bars, shown for a few of the data points, represent 1 standard deviation of the mean. The pressure of the argon gas in the second cell is measured using a barometer filled with diffusion-pump oil whose specific gravity is 1.07. The modulation of the ionization signal is clearly seen in the figure. The solid line is fitted to the data by adjusting the period, amplitude, average value, and phase. The depth of modulation (the ratio of the amplitude of the sine curve to the average value) is 15%. Deviation from the maximum possible value of 30% could be due to a number of factors such as imperfect beam overlap, nonoptimal ratio of the transition amplitudes at the focus (M), or saturation of the optical transition or ac Stark shifts of the $6p$ level leading to an intensity dependence other than I^5. Further work is required to sort these out. The pressure difference of the argon required to change the phase by 2π is approximately 6.1 ± 0.6 torr, with the uncertainty limited by slow drifts in the experiment. Using Eq. (2), this yields a refractive-index difference of $(4.6 \pm 0.4) \times 10^{-5}$, if we do not correct for variations in the temperature across the argon chamber. This is in reasonable agreement with extrapolations of refractive-index data reported previously[14] for argon, yielding $\Delta n = 4.5 \times 10^{-5}$.

In conclusion, we have observed an interference between two optical processes which can induce a transition between the same two atomic states. By changing the relative phase between the fields inducing these transitions we can modulate the interference from destructive to constructive. Future studies are planned to determine the maximum depth of modulation attainable, and to determine the utility of this effect in measuring the intensity dependence of nonlinear interactions. This work was supported by the National Science Foundation, Grant No. ECS-8451259. Support from Purdue University is also acknowledged.

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