Polarization dependence and interference in four-wave mixing with Rydberg levels in rubidium vapor

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We have observed an interference in four-wave mixing involving two Rydberg states that play the key role of near two-photon resonant intermediate levels. This interference, due to the coherence of the nonlinear process, can be explained either classically or quantum mechanically and it depends on the lasers' polarizations and detunings. We have performed calculations that are in very good agreement with the experimental data. Our results provide new information about the system, such as ionization rates of the Rydberg levels and the ratio of reduced matrix elements involved. [S1050-2947(98)12509-X]

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The study of interference effects in multilevel atomic systems has been an active field of research in recent years and has allowed the observation of a variety of physical phenomena in coherent and quantum optics [1]. Moreover, there has been considerable interest in the application of an ''interfering-pathways'' scheme for coherent laser control of chemical reactions [2]. In some cases, the interference can occur as a result of the generation of a new eletromagnetic field in the medium, through a nonlinear interaction, as in the cancellation of the multiphoton ionization due to the interference between three-photon excitation and third-harmonic generation [3]. In this phenomenon a difference of phase of 180° between the transition amplitutes leads to a destructive interference. Usually, the phenomena of interference are based on an induced phase relation between the fields involved in the different channels that connect the same initial and final states. This relative phase can be determined by the probability amplitudes corresponding to each path and by the polarizations of the driving fields. Using two fields with parallel polarizations, Bjorkholm and Liao [4] demonstrated fine-structure destructive interference on a resonant two-photon transition in sodium. Recently, Garret and Zhu [5] used an internally generated interfering-pathways technique to create a coherently controlled excitation in mixtures of Xe and Kr gases.

In this paper we report combined theoretical and experimental results on the dependence of the four-wave-mixing (FWM) signal with the polarizations of the two incident laser beams, using Rubidium as the nonlinear medium. By comparing the FWM spectra for different polarizations we have been able to observe an interference when two neighboring Rydberg levels, 16*d* and 18*s*, play the role of near two-photon resonant intermediate levels. The four-level scheme investigated in this experiment is similar to that of Xia and co-workers [1], but here the two upper levels are Rydberg atomic states. Also, the interference is observed in the FWM signal so it can be explained either classically or quantum mechanically. In the classical version, two polarizations are induced in the medium, with a phase coherence created by the FWM process. The quantum explanation is based on the fact that the signal can be generated via two separate indistinguishable quantum paths involving different Rydberg levels, resulting in an interference term. Moreover, theoretical analysis results in an excellent fit of the experimental data. The adjusted parameters give new information about the system and allow estimates of the ionization rates and ratio of the coupling's radial matrix elements involved.

The experimental setup is sketched in Fig. 1. A Nd:YAG laser (pulse duration 10 ns and repetition rate 5 Hz) is frequency doubled and is used to pump a dye laser with a wavelength of approximately 602 nm and a linewidth of 0.5 cm^{-1}. This laser, with frequency ω_1, can be tuned to resonance with two-photon transitions from the ground state, 5*s* to Rydberg levels ranging from 17*s* to 19*s* and 15*d* to 17*d*. For the present experiment, only levels 16*d* and 18*s* are relevant. Part of the fundamental mode (IR) of the Nd:YAG laser is combined with the dye laser and the two copropagat-

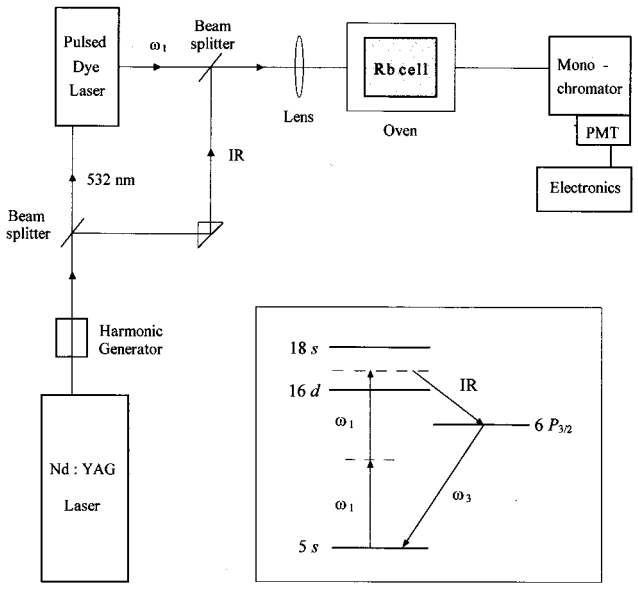

FIG. 1. Schematic view of the experimental apparatus with inset showing the relevant rubidium energy levels.

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ing beams are focused into a Rb vapor-cell heated to 180 °C. The IR beam, with fixed frequency ω_2, is quasiresonant with the transition from the Rydberg levels back to the $6P_{3/2}$ level (cf. inset in Fig. 1). A fourth photon is then generated at frequency $\omega_3=2\omega_1-\omega_2$ and brings the atoms back to the ground state. This fourth photon has a wavelength of approximately 420 nm and is analyzed in a monochromator and detected by a photomultiplier tube. The focusing of the two beams in the middle of the cell enables us to have good phase matching and a strong FWM generated signal is observed. Four-wave mixing spectra are obtained by measuring the intensity emitted as a function of the dye laser's frequency.

At first one would expect two peaks centered on the frequencies corresponding to resonant two-photon excitation of the 16*d* and 18*s* levels. Closer inspection reveals, however, a third peak at an intermediate frequency between the 16*d* and 18*s* peaks. This third peak is associated with the resonant three-photon transition from the ground state to the $6P_{3/2}$ state and the subsequent resonant emission back to the ground state. We will refer to it as the $6P_{3/2}$ peak. The three-photon transition from $5s$ to $6P_{3/2}$ has a two-photon contribution from the dye laser and one photon from the IR laser (cf. inset in Fig. 1). For this dye laser frequency, the 16*d* and 18*s* Rydberg levels, separated by $\approx 14.4\ \mathrm{cm}^{-1}$, contribute to the FWM signal in a quasiresonant way. As a matter of fact, both levels contribute significantly to the signal at all intermediate frequencies between the 16*d* and 18*s* peaks. Quantum mechanically, this means that there are two indistinguishable paths leading to the same final outcome and we expect interference between the probability amplitudes corresponding to each path. In particular, we expect this interference to be important for the dye laser frequency corresponding to the $6P_{3/2}$ peak.

As we noted before, this quantum mechanical explanation has, in this case, a classical counterpart. Classically, the FWM signal is due to polarizations created in the medium that generate the field at 420 nm. Thus, the $6P_{3/2}$ peak observed will reveal an interference between the fields induced by the two polarizations generated in the sample, at the same frequency but with different phases corresponding to the 16*d* and 18*s* levels. The coherence between these polarizations is guaranteed by the coherence of the FWM process itself.

It is not easy, in principle, to detect this interference because there are not that many parameters that can be varied. On the other hand, variations in the interference signal should be expected if one can change the relative contributions of the 16*d* and 18*s* levels. This can be done by changing the polarization of the dye laser. For a linear polarization we expect to have similar contributions from both Rydberg levels. However, if the dye laser is circularly polarized, selection rules forbid two-photon excitation from the ground state to the 18*s* level. In this situation, there is no longer any interference because only one quantum path is accessible to the system. Even for dye laser linear polarization we will see below that the interference can vary from destructive to constructive depending on its polarization relative to the IR laser's fixed linear polarization.

We have developed a simple model to understand our spectra based on the density matrix formalism applied to a four-level system. The density matrix operator obeys Liouville's equation

$$\frac{\partial\hat{\rho}}{\partial t}=-\frac{i}{\hbar}[\hat{H},\hat{\rho}]+(\text{relaxation terms}). \tag{1}$$

In order to avoid writing long subscripts we note the levels $|5s\rangle$, $|6P_{3/2}\rangle$, $|16d\rangle$, and $|18s\rangle$, respectively, $|a\rangle$, $|b\rangle$, $|c\rangle$, and $|d\rangle$. The interaction Hamiltonian's matrix elements are then given by ($i=c$ or d)

$$V_{ia}=\frac{K_{ia}E_1^2}{\hbar};\quad V_{ib}=-\frac{\mu_{ib}E_2}{\hbar}, \tag{2}$$

where the effective two-photon coupling constant K_{ia} is

$$K_{ia}=\frac{1}{\hbar}\sum_j\frac{\mu_{ij}\mu_{ja}}{\omega_{ij}-\omega_1}, \tag{3}$$

and μ_{ij} is the electric dipole matrix element between levels i and j.

The FWM signal intensity is proportional to the polarization induced in the medium at the frequency $2\omega_1-\omega_2$ determined by the density matrix element ρ_{ab},

$$I_{FWM}\propto|P^{(3)}(2\omega_1-\omega_2)|^2\propto|\rho_{ab}|^2. \tag{4}$$

Since there are two possible quantum paths, involving one or the other Rydberg level, the steady-state solution for this density matrix element will involve a sum of probability amplitudes corresponding to each level. Applying perturbation theory, up to second order in E_1 and first order in E_2 [6], we obtain

$$\begin{aligned}|\rho_{ab}|^2&=|\rho_{ab}^{(c)}+\rho_{ab}^{(d)}|^2\\&=\frac{|V_{ad}|^2|V_{db}|^2}{(\gamma_{ab}^2+\Delta_2^2)}\left\{\frac{1}{[\gamma_{ad}^2+(\Delta_1')^2]}+\frac{\mathcal{R}^2}{(\gamma_{ac}^2+\Delta_1^2)}\right.\\&\quad\left.+\frac{2\mathcal{R}(\gamma_{ad}\gamma_{ac}+\Delta_1\Delta_1')}{[(\gamma_{ad}\gamma_{ac}+\Delta_1\Delta_1')^2+(\gamma_{ad}\Delta_1-\gamma_{ac}\Delta_1')^2]}\right\},\end{aligned} \tag{5}$$

where

$$\mathcal{R}=\frac{V_{ac}V_{cb}}{V_{ad}V_{db}}. \tag{6}$$

In Eq. (5) we have defined the detunings $\Delta_1=2\omega_1-\omega_{ac}$, $\Delta_1'=2\omega_1-\omega_{ad}$ and $\Delta_2=2\omega_1-\omega_2-\omega_{ab}$, and introduced as well the relaxation rates γ_{ab}, γ_{ac}, and γ_{ad}. Fine and hyperfine structures and the Doppler broadening, which are unresolved by the lasers, are neglected and we consider that only the $5p$ state contributes to the two-photon transitions between the ground state and the Rydberg levels.

This simple theory can be readily applied to predict the shape and dependence of the FWM signal as a function of the dye and IR polarizations. In order to do so, it is necessary to take into account the Clebsch-Gordan coefficients when calculating the effective couplings. Actually, some of the constants defined above are not *a priori* so well known in our system. For example, the relaxation rates of the Rydberg levels and even of the $6P_{3/2}$ state are not purely radiative. As

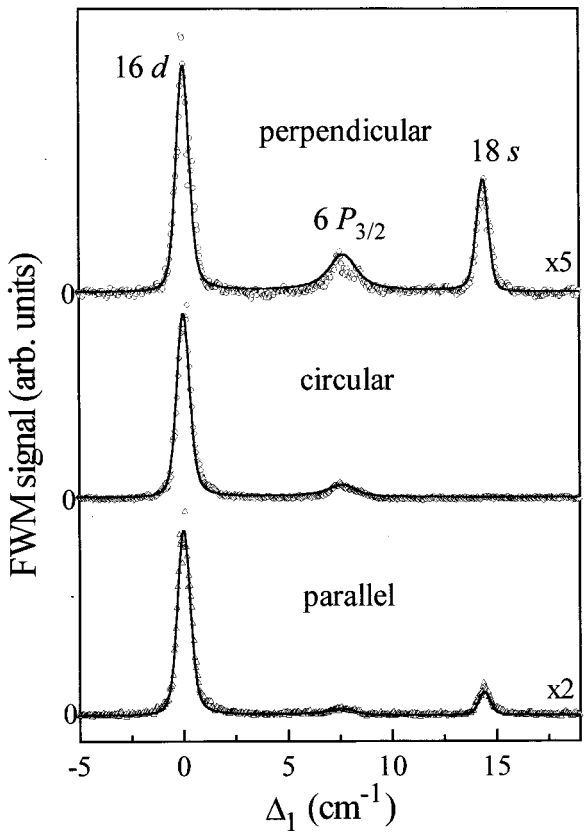

FIG. 2. Four-wave-mixing intensity as a function of the detuning Δ_1 for different dye laser polarizations. Note that the signals for parallel and perpendicular polarizations are multiplied, respectively, by 2 and 5. Dye laser and IR laser intensities were $I_{\mathrm{dye}}=1\times10^{8}$ W/cm^2, $I_{\mathrm{IR}}=3\times10^{9}$ W/cm^2. The atomic density was $n=4.4\times10^{14}$ atoms/cm^3. The solid lines are theoretical fits based on Eq. (5) with the respective Clebsch-Gordan coefficients.

a matter of fact, the three levels can be ionized directly by one- and two-photon transitions induced by the dye and IR lasers. Instead of estimating the ionization cross sections in each case, we prefer to use Eq. (5) leaving these parameters (as well as the reduced matrix elements) free to fit the experimental data.

Our experimental results were obtained for three different dye laser polarizations, always maintaining a fixed linear IR laser polarization and assuming the axis of quantization in the direction of the propagating beams. The three polarizations are linear parallel to the IR polarization, linear perpendicular to the IR polarization, and circular polarization. We shall refer to them simply as parallel, perpendicular, and circular. Experimentally we can switch from one configuration to the other by means of a half-wave plate and a Fresnel rhomb situated in the dye laser beam path. The FWM spectra obtained for each polarization represent an average over four independent scans of the dye laser in order to minimize noise. The results are depicted in Fig. 2, together with theoretical fits corresponding to each case. We consider that γ_{ab} is given by the width of the $6P_{3/2}$ peak (≈1.5 cm^{-1}), which is limited by the linewidth of the IR beam. To take into account the finite linewidth of the dye laser, we perform a convolution with its measured Gaussian line shape. The theoretical curves are then obtained by numerical integration of Eq. (5). An excellent agreement between theory and experiment is clearly observed. The fit is actually made only for one polarization and the same parameters are used to compare the theory with the data for the other two polarizations. This proves the consistency of the whole treatment. The data we used to perform the fit was for perpendicular polarization because, in this case, all three peaks have comparable intensities.

The parameters ajusted are $\gamma_{16d}=(0.252\pm0.008)$ cm^{-1}, $\gamma_{18s}=(0.178\pm0.010)$ cm^{-1} and the ratio of the reduced matrix elements $R=5.1\pm0.5$, given by

$$R=\frac{\langle 5p||r||16d\rangle\langle 16d||r||6p\rangle}{\langle 5p||r||18s\rangle\langle 18s||r||6p\rangle}. \tag{7}$$

The ratio R reflects the spatial overlap of the $5p$ and $6p$ levels' wave functions with the excited $18s$ and $16d$ levels. This value for R can be compared with a numerical calculation using a method based on the diagonalization of the energy matrix [7]. For the levels of Rb involved we obtain R = 4.2, in relative agreement with the fitted value, thus showing the consistency of our analysis based on the four-level system. In this model we have not considered absorption effects. In fact, we expect a stronger absorption when the dye laser is tuned to the $6P_{3/2}$ three-photon transition than when either of the Rydberg states are two-photon resonant. This may lead to a decrease of the fitting value of R and a better agreement with the numerical calculation. The values for the relaxation rates indicate that ionization is the dominant mechanism [8]. Futhermore, from these values we can estimate photoionization cross sections, at the dye laser frequency (≈0.15 Ry above threshold), of $\sigma_{16d}=25\times10^{-18}$ cm^2 and $\sigma_{18s}=17\times10^{-18}$ cm^2, with an error of 30% due to the uncertainty in the dye laser intensity. Note also that $\sigma_{16d}>\sigma_{18s}$, which is not surprising if we compare these with the photoionization cross sections obtained for lower-lying states [9] and theoretical calculations [10] at threshold, for $n<20$.

In principle, the interference effect could be seen by simply comparing the intensities corresponding to the $6P_{3/2}$ peak for the three different polarizations. The intensity of each peak should reflect the probability of observing that peak for the given experimental conditions. However, as we can observe in the spectra of Fig. 2 the whole signal changes in intensity when we switch from one polarization to another. So, in order to compare the relative intensities of the $6P_{3/2}$ peak, it is necessary to somehow normalize it. We have chosen to divide its intensity by the sum of the intensities of the $16d$ and $18s$ peaks in each case. We can call this quantity I_{rel} and express it as

$$I_{\mathrm{rel}}=\frac{I(6P_{3/2})}{I(16d)+I(18s)}. \tag{8}$$

Our results give $I_{\mathrm{rel}}^{(\parallel)}=0.04$, $I_{\mathrm{rel}}^{(\mathrm{circ})}=0.09$, and $I_{\mathrm{rel}}^{(\perp)}=0.14$. We are therefore observing destructive interference for parallel polarization, constructive interference for perpendicular polarization and no interference (as expected) for circular polarization. One should note that the value obtained for circular polarization lies exactly in the middle of the other two. This indicates a way to observe interference *fringes* in this system. All that is necessary is to measure I_{rel} for several different dye laser *linear* polarizations.

These results agree with the predictions based on Eq. (5) taking into account the appropriate Clebsch-Gordan coefficients. Actually, for perpendicular polarization the interference is constructive when twice the frequency of the dye laser is tuned in between the two Rydberg levels and destructive outside; while for parallel, the opposite is true. This behavior, involving three-photon absorption, is similar to what happens in two-photon transition interference [4]. We can also use the quantity P_L [11], defined as

$$P_L = \left(\frac{I_{\mathrm{rel}}^{(\parallel)} - I_{\mathrm{rel}}^{(\perp)}}{I_{\mathrm{rel}}^{(\parallel)} + I_{\mathrm{rel}}^{(\perp)}} \right), \tag{9}$$

to measure an interference ''contrast.'' For the present experiment P_L is of order of -56%. Measurements of P_L from $+100\%$ to -100% have been obtained in two-photon, two-color polarization experiments in sodium [12] and rubidium [11,13]. In this case, when $P_L = -100\%$, the interference between the two paths for the two-photon excitation is responsible for the previously reported two-photon transparency [4]. In our case, if we tuned the IR laser (or if we used the dc Stark effect to shift the Rydberg levels) we should be able to cancel the $6P_{3/2}$ peak, in the parallel polarization spectrum, for $\Delta_1/\Delta_1' = -\mathcal{R}$ and $\Delta_2 = 0$. The maximum of destructive interference will occur over the $6P_{3/2}$ peak resulting in a three-photon transparency.

In conclusion, we have observed constructive and/or destructive three-photon interference in FWM with Rydberg levels in Rb vapor and we have extracted new information about the system from our data. Future experiments to observe the same kind of interference in multiphoton ionization in this system are presently being prepared. In this case, the nature of the interference will be purely quantum mechanical.

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