Competition between quantum beatings at 608 cm^{-1} and 70 cm^{-1} in Rb observed by Parametric Four-Wave Mixing

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Abstract: The competition between quantum beatings at 608 cm^{-1} and 70 cm^{-1} in Rb is detected and controlled by using Parametric Four-Wave Mixing. The Rb number density, laser intensity and the laser polarization control the competition.

OCIS codes: (020.1670) Coherent optical effects; (190.4380) Nonlinear optics; four-wave mixing

Quantum beating may originate from the interference of wave packets, and can be observed by pump-probe experiments in which wavepackets are interrogated by parametric four-wave mixing (PFWM). Quantum beatings at 608 cm^{-1} and 70 cm^{-1} in Rb have been observed simultaneously by detecting an axially phase-matched PFWM signal, a beam of coherent radiation at a wavelength of 420 nm. Quantum beating at 608 cm^{-1} in Rb stems from the coherent two-photon excitation of the 7s and 5d states, and the energy defect between them (7s-5d $_{(J=5/2)}$), 608 cm^{-1}, is embedded into the FWM signal. Quantum beating at 70 cm^{-1} in Rb, however, originates from the sequential excitation from 5s (ground) via the 5p state to the 5d state, and the energy difference ($5d_{(J=5/2)}$-$5p_{(J=3/2)}$)-($5p_{(J=3/2)}$-5s), 70 cm^{-1}, is also superimposed onto the FWM signal. A broad bandwidth laser pulse thus serves two purposes. The first is to coherently excite the 7s and 5d states in Rb, producing a wave packet comprising both states. The second is to sequentially excite Rb atoms from 5s to 5p and then from 5p to 5d, producing a wave packet comprising the 5d, 5p and 5s states. A pump-probe configuration, combined with PFWM, has the advantages of fs temporal resolution and a detection mechanism (coherent radiation at the signal frequency) that offers an extraordinary S/N ratio. The intensity of the PFWM signal at 420 nm is recorded as a function of the time delay between the pump and probe pulses. Analyzed by the Time Dependent Fourier Transform, the data in the time domain are transformed into the frequency domain, giving rise to the temporal evolution of the amplitude of the quantum beats at 608 cm^{-1} and 70 cm^{-1}.

Several factors influencing the competition are analyzed and then exploited to actively control the competition between quantum beating at 608 cm^{-1} and 70 cm^{-1}. The first is the Rb number density. The 5d state is simultaneously involved in two parametric FWM processes: $5^2S_{1/2}\rightarrow\rightarrow 5^2D_{5/2}\rightarrow 6^2P_{3/2}\rightarrow 5^2S_{1/2}$ (channel 1), and $5^2S_{1/2}\rightarrow 5^2P_{3/2}\rightarrow 5^2D_{5/2}\rightarrow 6^2P_{3/2}\rightarrow 5^2S_{1/2}$ (channel 2), both of which contribute to the FWM signal. Fig. 1 shows the temporal evolution of the amplitudes of quantum beat at 608 cm^{-1} and 70 cm^{-1} for Rb number density of (a) 6×10^{13} cm^{-3} and (b) 3×10^{14} cm^{-3}. Quantum beating at 608 cm^{-1} is weaker or stronger than that at 70 cm^{-1} because phase matching alternately favors a channel and, hence, suppresses the other. The second is the intensity of the fundamental optical field. In the perturbative view, the probability of a two-photon transition is proportional to the square of the intensity of the fundamental optical field, providing another means for controlling the competition (see Fig. 2). The third is the polarization of the fundamental optical field. Considering the dependence of selection rules on polarization for one and two-photon transitions, one can also control the competition by using pump pulses with different polarizations, which is shown in Fig. 3. All of these effects will be demonstrated and discussed from the viewpoint of utilizing atomic wavepackets as detectors for molecular spectroscopy.

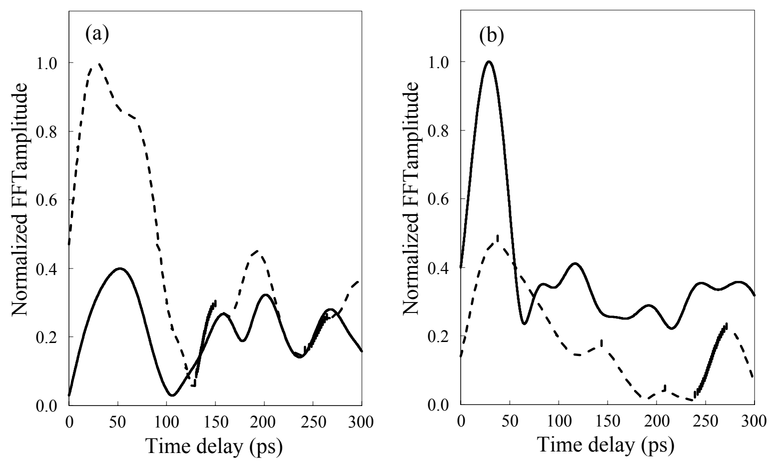

Fig. 1. Variation with time delay of the Fourier amplitude of the 608 cm^{-1} (solid curve) and 70 cm^{-1} (dashed curve) frequency components of Rb wavepackets. In taking these data, the pump is parallel to the probe, and both are linearly polarized. The Rb number density is (a) 6×10^{13} cm^{-3} and (b) 3×10^{14} cm^{-3}.

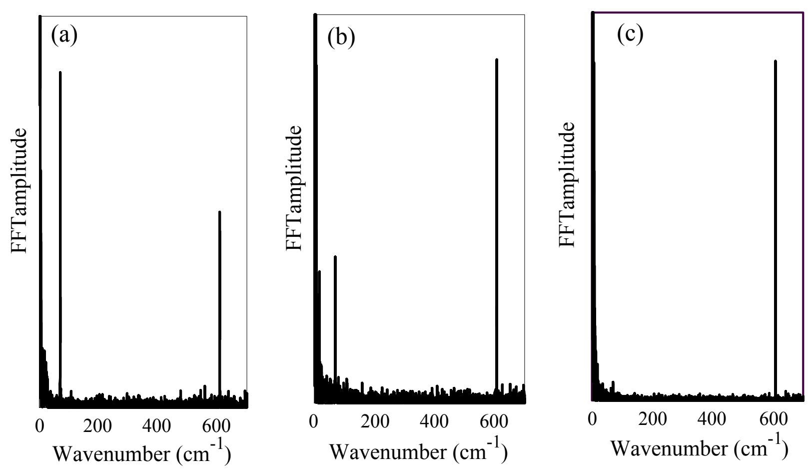

Fig. 2. FFT amplitudes for a fixed Rb number density (6×10^{13} cm^{-3}). The pump is parallel to the probe, and both are linearly polarized. The laser pulse width is 100 fs, the repetition rate is 1 kHz and the total peak intensity of the pump and probe pulses is (a) 2.5×10^{11} Wcm^{-2}, (b) 5.5×10^{11} Wcm^{-2} and (c) 8.5×10^{11} Wcm^{-2}.

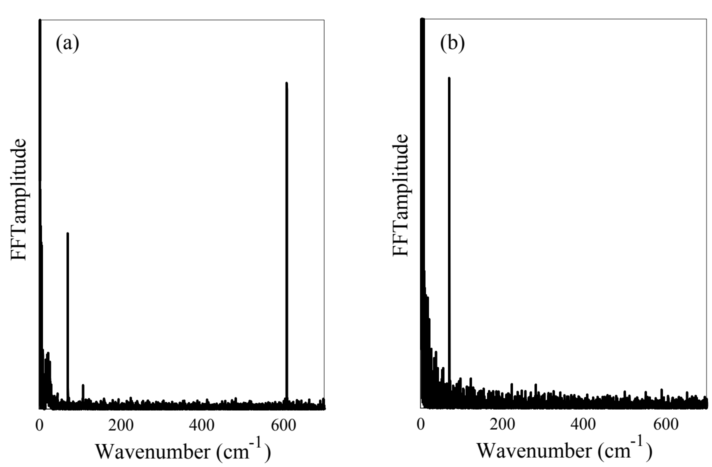

Fig. 3. FFT amplitudes for a fixed Rb density (6×10^{13} cm^{-3}). The total peak intensity of the pump and probe pulses is 2.5×10^{11} Wcm^{-2}. (a). The pump is parallel to the probe, and both are linearly polarized. (b). Both the pump and probe are circularly polarized.