Interference between overlapping resonances: A route to coherent control of resonance lifetimes and photofragment distributions in the weak-field limit

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Synopsis Two different coherent control schemes in the weak-field regime are discussed. They are based on the manipulation of the mechanisms of interference between overlapping resonances when they are simultaneously populated in a coherent superposition. With the first control scheme, enhancement by a factor of three of the lifetime of a specific single resonance state of the superposition created, is achieved. The second control scheme allows one to control the transient photofragment distributions by means of pure phase modulation of the pump laser pulse for a long time window after the pulse is over.

Overlapping resonances possess two very interesting features: (1) They are nonorthogonal states that can interfere between themselves; (2) they have a finite lifetime that can vary remarkably for different resonances. The combination of these two features makes possible a rather flexible control of the mechanisms of interference when the overlapping resonances are populated simultaneously in a coherent superposition. Thus, preparing such a superposition provides a powerful tool to design and develop coherent control schemes.

It has been recently shown that the lifetime of an overlapping resonance strongly depends on the amplitudes of the other overlapping resonances populated in a superposition [1]. This is the basis of a control scheme which, in its simplest version, creates a superposition of two overlapping resonances in the complex Ne-Br_2(B,v'=27), using one pump laser pulse to excite each of the two resonances. By varying the delay time and the ratio of amplitudes between the two pump pulses, it is possible to control the amount of population that is simultaneouly excited to both resonances, and therefore to control the intensity of interference between them [2,3]. In this way, an enhancement by a factor of three of the lifetime of one of the resonances is achieved (see Figure 1) [2].

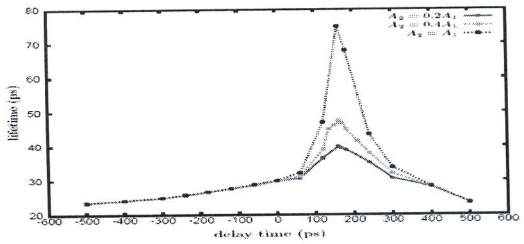

Figure 1. Resonance lifetimes vs delay time and three different ratios of amplitudes between pulses.

In the second control scheme, a single fixed bandwidth pump pulse is used to excite the same superposition of two resonances in Ne-Br_2(B,v'=27). By applying different linear chirps to the pulse, interference between the resonances can be controlled. As a result, phase effects of increasing intensity are produced in the transient vibrational populations of the Br_2(B,v_f<v') fragment as the chirp rate β_0 increases (see Figure 2) [4]. These pure pulse phase modulation effects occur for a long time window (about 200 ps) after the pulse is over.

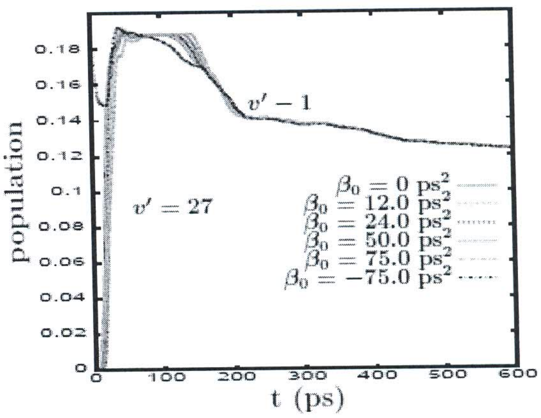

Figure 2. Transient Br_2(B,v_f=v'-1) fragment vibrational populations using different pulse chirps.

References

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