Ultranarrow resonance due to coherent population oscillations in a Λ-type atomic system

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It is well known that ultranarrow electromagnetically induced transparency (EIT) resonances can be observed in atomic gases at room temperature. We report here the experimental observation of another type of ultranarrow resonance, as narrow as the EIT ones, in a Λ system selected by light polarization in metastable 4He at room temperature. It is shown to be due to coherent population oscillations in an open two-level system (TLS). For perpendicular linearly polarized coupling and probe beams, this system can be considered as two coupled open TLSs, in which the ground-state populations exhibit antiphase oscillations. We also predict theoretically that in the case of two parallel polarizations, the system would behave like a closed TLS, and the narrow resonance associated with these oscillations would disappear.

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Coherent population oscillations (CPO) and electromagnetically induced transparency (EIT) are two phenomena that can give rise to resonances much narrower than the relaxation rate of optical coherences. The former happens in two-level atomic systems (TLS), when the beat note between a coupling beam and a coherent probe beam leads to a temporal modulation of the population difference. The width of the induced transparency window is then limited by the population modulation of the population difference. The width of the coupling beam and a coherent probe beam leads to a temporal relaxation rate of optical coherences. The former happens in netically induced transparency (EIT) are two phenomena within the order of 100 µs, leading to an effective Raman coherence lifetime of the upper-level population lifetime, EIT usually leads to the narrowest resonances. In the last two decades, such phenomena have been observed for light storage experiments [7–10].

In this Rapid Communication, we report the experimental observation of an ultranarrow resonance in the absorption spectrum of a hot atomic vapor. This resonance, which cannot be attributed to EIT, is shown theoretically and experimentally to be due to CPO in the two coupled TLSs provided by the Λ system. The two ground-state populations exhibit antiphase oscillations, while the total population is conserved. We compare all the features of this CPO resonance with those of the EIT ones.

The experiment uses metastable helium at room temperature. The 2S1/2 → 2P1/2 transition permits us to isolate a pure Λ system involving only electronic spins and in which the Raman coherence lifetime is limited by the transit time of the atoms through the laser beam [11]. This transit time is lengthened thanks to nondephasing collisions with ground-state atoms, leading to an effective Raman coherence lifetime of the order of 100 µs [12]. Figure 1(c) gives the schematic of the experimental setup. The helium cell, filled with 1 Torr of 4He, is 6 cm long and has a diameter of 2.5 cm. It is placed inside a three-layer μ-metal shield for isolation from magnetic field inhomogeneities. The coupling and probe beams are derived from the same laser diode at 1.083 μm. The beam diameters are about 1 cm inside the cell. Helium atoms are excited to the metastable state by a rf discharge at 27 MHz. The coupling and probe beams are controlled in frequency and amplitude by two acousto-optic modulators (AOMs), and recombined with a polarizing beam splitter (PBS). The probe power is about 50 µW and the coupling power can be varied between 0.5 and 22 mW. A quarter-wave plate (λ/4) located at the entrance of the cell lets us alternate between orthogonal circular (σ ⊥) and linear (lin lin) polarizations. A variable longitudinal magnetic field (B) generated by a solenoid surrounding the helium cell lifts the degeneracy of the lower sublevels. The Landé factor is 2 for the ground state, leading to Zeeman shifts of ±2.8 kHz/mG for the 2S1/2, m = ±1 levels. After the cell, polarization optics allows detection of only the probe.

In the usual configuration for EIT experiments along the 2S1/2 → 2P1/2 transition in 4He* [12], one uses circular polarizations for the pump and probe beams. Since the m = 0 → m = 0 transition is forbidden, a σ+ coupling beam pumps the atoms into the m = +1 ground-state sublevel, which is probed by a σ− beam [see Fig. 1(a)]. In contrast, when the coupling beam is linearly polarized, atoms are equally pumped into both m = ±1 sublevels, which can then be probed by a perpendicular linearly polarized probe beam [see Fig. 1(b)]. The experiments and simulations presented here compare these σ ⊥ σ and lin ⊥ lin configurations.

In the σ ⊥ σ configuration, Fig. 2(a) reproduces the experimentally recorded transmission spectrum of the probe versus δ with and without a magnetic field. As expected, the Zeeman shift ΔZ induced by the magnetic field shifts the EIT resonance. The Raman resonance is obtained for δ = ωC − ωP = 2ΔZ, which is in perfect agreement with our data. Now, Fig. 2(b) shows the results from the same experiment except that the pump and probe beams are now linearly and orthogonally polarized (lin ⊥ lin configuration). Thus, each beam equally excites the transitions Δm = ±1, as seen from...
The unexpected resonance mentioned in the text refers to a phenomenon where the central resonance does not correspond to any Raman resonance involving both optical transitions. It occurs when the coupling and probe beams have the same frequency, while both transitions experience opposite frequency shifts induced by the applied magnetic field. The appearance of such a peak in the context of an EIT experiment is thus quite surprising.

The fact that this unexpected resonance occurs at zero frequency difference \( \delta \) between the pump and the probe is reminiscent of CPO. However, the width of usual CPO resonances is given by the decay rate of the population of the upper level. In our case, since the lifetime of the excited level is of the order of 100 ns, this would lead to a resonance width of the order of 1 MHz. It is clear from Fig. 2(b) that the width of this extra central resonance is in the kHz range, and is quite close to the width of the side EIT resonances in Fig. 2(b) or of those in Fig. 2(a). We have also checked that EIT involving dressed states in a two-level system gives rise to resonances whose widths are much larger than those observed here. Consequently, the mechanism described in Ref. [13] is not relevant here. Thus, if we want to interpret this extra resonance as due to population oscillations, it has to be related to the lifetime of the lower level, which in our case is limited by the transit time and is thus compatible with the observed widths.

A simple and effective way to check whether a resonance is due to population oscillations or not is to observe whether the introduction of a dephasing effect, which decreases the lifetime of the coherences, affects it or not. With this aim, we record the evolution of the widths of the different resonances with the coupling intensity, in the presence of an inhomogeneous magnetic field, simply realized by taking the cell partially out of the magnetic field shielding, and then again with proper magnetic shielding. The result is reproduced in Fig. 3(a). The standard EIT resonance is of course extremely sensitive to magnetic field gradients, because it relies on the Raman coherence. In contrast, the central resonance in the \( \perp \perp \) configuration here is totally unaffected by these gradients. This proves that this resonance is governed by the population lifetime in the lower state of the transition, and not by the Raman coherence lifetime.

We thus see a resonance due to population oscillations in the ground state. This resonance is observable here, and never in the case of CPO in a standard closed TLS, due to the fact that our three levels constitute two open TLSs. The extra resonance we get is thus linked to the extra resonances predicted by Friedmann et al. [14] in 1986 in the case of four-wave mixing (FWM), when an intermediate decaying state is added to a TLS.

We could reproduce the experimental results by performing a first-order Floquet expansion of the density matrix of the three-level system in a manner similar to Wong et al. [15]. Such simulated spectra are shown in Fig. 3(b) for two values of the Raman coherence linewidth. These confirm that the central resonance is not linked to the Raman coherences, unlike the EIT side peaks.

If the central resonance is, as we suspect, due to population oscillations in the lower state sublevels, we should be able to reproduce the experimental results in the rate equation approximation, i.e., with all the coherences adiabatically
discussed both theoretically and experimentally by different groups at the end of the 1980s and the beginning of the 1990s [17,18], with an emphasis on FWM, considerably different from the point of view developed here. Figure 4(d) proves that the narrow dip of Fig. 4(b) is due to a decrease of the amplitude of the oscillations of the lower-level population.

However, our Λ system is more than an open TLS: it consists of two open TLSs which are interdependent and excited by the same pair of coupling and probe fields. One could thus expect the lower-level CPO of these two TLSs to interfere constructively or destructively, depending on the relative signs of their excitation fields. This is why the results of Fig. 5 permit us to compare the results of the Floquet simulations in our Λ system for perpendicular (lin ∥ lin) or parallel (lin || lin) linearly polarized pump and probe beams. Figure 5(a) shows that the ground-state CPO resonance is present only in the lin ∥ lin configuration, but disappears in the lin || lin configuration.

Indeed, in the lin ∥ lin configuration [see Fig. 5(b)], the populations of the two ground-state sublevels oscillate in antiphase, due to the fact that they are driven by intensity modulations which are in antiphase. The two CPO resonances induced by the two legs of the Λ, then add constructively, giving birth to a sharp resonance limited by the decay rate of the populations of the ground states. In contrast, in the lin || lin configuration, the populations of the two ground states oscillate in phase [see Fig. 5(c)], and the system behaves like a closed TLS, showing no resonance in the kHz range. Of course, as shown in Fig. 5(d), the system can still exhibit the much broader, usual CPO resonance with a width given by Γ0, just like an ordinary closed TLS.

Notice that the values of the parameters used in the simulations shown in Figs. 3–5 correspond to the experimental values.
The oscillating part in the linear configuration. (c), (d) Evolutions versus \( \delta \) of the oscillating parts \( \Delta N_{g1} = (N_{g1} e^{-i\delta} + c.c.) \) of the excited state (continuous purple), and \( \Delta N_{g2} = (N_{g2} e^{-i\delta} + c.c.) \) (dotted green) of the two ground states, at a fixed time \( t \) in the linear configuration. (c), (d) Evolutions versus \( \delta \) of the oscillating part \( \Delta N_{e} \) of the excited state (continuous purple) and of the sum \( \Delta N_{g1} + \Delta N_{g2} \) of the oscillating parts of the two ground-state populations (dashed blue), in the linear configuration.

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However, these papers focused on either electromagnetically induced absorption [19,20] or spectral analysis [21] and did not discuss the physics underlying the phenomenon or gave ambiguous explanations mixing Zeeman coherences and oscillations [22,23].

The interest in CPO has lasted for more than a decade since many people work on possible applications of slow and fast light, in particular, for microwave photonics [24–27]. In addition, recent theoretical proposals suggest the use of such long-lived CPOs even for applications in spatial optical memories [28] or narrowband biphoto generation [29]. The system described here is of interest as it can be made to behave as either a closed or an open TLS by a simple change of the polarization direction of one of the coupling and probe beams. It also has the advantage of exhibiting conservation of its total population, avoiding the need for a repumping laser. Moreover, the decay rate of the ground-state population is very long and is not limited by spontaneous emission, but only by the transit of the atoms through the laser beam. Other A systems in solids (such as rare-earth ions or nitrogen-vacancy centers in diamonds) might even go beyond the present limitation set by the transit time. Such coupled open TLSs are thus good candidates for the experimental implementation of the recent theoretical proposals based on narrow CPO effects.