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Dicke coherent narrowing in two-photon and Raman spectroscopy of thin vapor cells

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The principle of coherent Dicke narrowing in a thin vapor cell, in which sub-Doppler spectral line shapes are observed under a normal irradiation for a $\lambda/2$ thickness, is generalized to two-photon spectroscopy. Only the sum of the two wave vectors must be normal to the cell, making the two-photon scheme highly versatile. A comparison is provided between the Dicke narrowing with copropagating fields, and the residual Doppler broadening occurring with counterpropagating geometries. The experimental feasibility is discussed on the basis of a first observation of a two-photon resonance in a 300-nm-thick Cs cell. Extension to the Raman situation is finally considered.

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Linear spectroscopy in a macroscopic low-pressure gas is usually sensitive to the Doppler broadening, as governed by $|\vec{k}|u$ (with u the most probable thermal velocity, and $\vec{k}$ the wave vector of the irradiating field). In cw multiphoton spectroscopy, when several irradiating beams $(\omega_i,\vec{k}_i)$ are used, the Doppler broadening generalizes to $|\Sigma\vec{k}_i|u$ [1]. When a cell of dilute gas is so thin that atomic (or molecular) trajectories are mostly from wall to wall, sub-Doppler spectral line shapes can be obtained in a variety of processes (optical pumping, etc.) under *normal* incidence; this results from a specific enhancement of the slow atoms contribution (see [2] and references therein), with respect to the transient nature of the atom-light interaction. In linear optical absorption, a particular coherent narrowing has been experimentally observed for a $\lambda/2$ cell thickness [3,4], analogous to the one mentioned by Romer and Dicke [5] in the microwave domain. The purpose of this paper is to show that the main features of this coherent Dicke narrowing can be extended to two- or multiphoton transitions, as long as an effective two-level model can be considered, and for a *normal* incidence condition now referring to the sum of photon wave vectors $|\Sigma\vec{k}_i|$.

Let us recall that for a cell filled with two-level atoms, the one-photon resonant transmission (or reflection [6,7]) behavior depends on the field radiated by the induced macroscopic polarization, resulting from the spatial summing of the local optical coherence, as given by the quantity $\mathrm{Re}[I_f]$ with

$$I_f = i\int_0^{L} \sigma_{21}(z)dz = i\int_{-\infty}^{+\infty}\int_0^{L} \sigma_{21}(z,v_z)f(v_z)dv_z dz. \quad (1)$$

In (1), L is the cell length, Oz is the axis normal to the thin cell, $f(v_z)$ the velocity distribution, usually assumed to be Maxwellian. Assuming a linear regime of interaction, $\sigma_{21}(z,v_z)$ is given as a result of the elapsed time $t=z/v_z$ from the wall departure (for $v_z>0$), where $\sigma_{21}=0$, by

$$\sigma_{21}(z,v_z) = \frac{i\Omega}{2D_{21}}[1-\exp(-D_{21}z/v_z)] \quad (2)$$

In (2), Ω is the Rabi frequency, and $D_{21}=\gamma_{21}-i(\Delta-\vec{k}\cdot\vec{v})$ characterizes the resonance with γ_{21} the relaxation coefficient of the optical dipole and Δ the frequency detuning between the field and the $|1\rangle\to|2\rangle$ transition. Note that assuming a normal incidence, the Doppler shift has been counted on the same direction as the one along which the interaction time is measured (i.e., for $\vec{k}\cdot\vec{v}=kv_z$). For $v_z<0$, z/v_z is replaced by $(z\text{-}L)/v_z$. The transmission line shape strongly depends on the L thickness [3,4,8]. It oscillates, with a pseudoperiodicity λ, from a sub-Doppler width (for $L=(2n+1)\lambda/2$) to a Doppler-broadened one (for $L=n\lambda$). The narrowest line shape is for the $\lambda/2$ thickness as in the situation analyzed by Romer and Dicke [5]. It exhibits a logarithmic singularity (in the limiting case $ku \gg \gamma_{21}$), originating in the $1/v_z$ contribution of fast atoms. Also, in spite of a mismatching brought by the Doppler detuning [3,4], all velocity groups interfere constructively up to the $\lambda/2$ thickness.

In the extension to a two-photon situation (see Fig. 1), one monitors the $(\omega_2,\vec{k}_2)$ probe beam transmission change induced by a $(\omega_1,\vec{k}_1)$ pump, governed by an integrated optical coherence $I_f^{2-\mathrm{ph}}=i\int_0^L\sigma_{32}(z)dz$. In an effective two-level model situation, i.e., when the irradiating frequencies ω_1 and ω_2 are far enough from the resonances ω_{21} and ω_{32}, the pump-induced optical coherence $\sigma_{32}(z)$ simply results from the transient regime of the resonant two-photon coherence

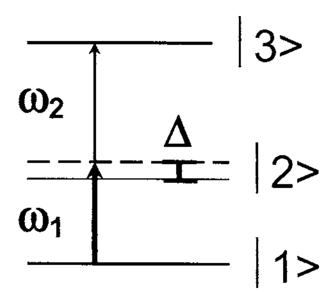

FIG. 1. A schematic of the atomic three-level system

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$\sigma_{31}(z)$. The assumption is that the sum of the exciting frequencies $\omega_1+\omega_2$ is nearly two-photon resonant, i.e., that $\Delta_{31}=(\omega_1+\omega_2)-\omega_{31}$ remains comparable with the two-photon width (homogenous width γ_{31}, or at least with the Doppler-broadened width $|\vec{k}_1+\vec{k}_2|u$), while the individual excitation frequencies ω_1 and ω_2 are largely detuned from the one-photon resonances ω_{21} and ω_{32}. In this situation, saturation effects are most often negligible and a third-order perturbation expansion (second order with respect to the "pump" field, first order with respect to the detected "probe" field) is sufficient to evaluate the atomic response σ_{32}.

The essence of the extension of the coherent Dicke narrowing to two-photon transition lies in the fact that the $\sigma_{32}(z,v_z)$ behavior is strictly analogous to the one of $\sigma_{21}(z,v_z)$ in (1), with the one-photon $\vec{k}$ vector simply replaced by the two-photon wave vector $\vec{k}_1+\vec{k}_2$. Namely, one shows (for $v_z>0$) that

$$\sigma_{32}(z,v_z) \approx \frac{i\Omega_1^2\Omega_2}{8\Delta^2 D_{31}}[1-\exp(-D_{31}z/v_z)] \tag{3}$$

[and z/v_z replaced by $(z-L)/v_z$ for $v_z<0$]. In (3), $D_{31}=\gamma_{31}-i(\Delta_{31}-(\vec{k}_1+\vec{k}_2)\cdot\vec{v})$ characterizes the two-photon resonance, while Δ is the detuning between the pump laser frequency and the one-photon excitation, and Ω_1 and Ω_2 are the respective Rabi frequencies associated to the pump (ω_1) and to the probe (ω_2) beams. In the vicinity of the two-photon resonance, the large value of Δ implies that $\Delta=\omega_1-\omega_{21}\approx\omega_{32}-\omega_2\equiv-\Delta_2$.

A justification for (3) can be obtained from an exact calculation of σ_{32}, as provided in the appendix in the nonrestrictive frame of a conservative three-level system. The full analogy of (3) with (2) is responsible for the two-photon coherent Dicke narrowing, but it should be noted that the complex phase factor now depends upon the *vectorial sum of the wave vectors* $\vec{k}_1+\vec{k}_2$. As a consequence, the width evolves from a Doppler broadening $|\vec{k}_1+\vec{k}_2|u$ to a sub-Doppler structure depending on γ_{13}, and the maximal narrowing is hence obtained for a thickness $L=\Lambda/2$ with $\Lambda=2\pi/|\vec{k}_1+\vec{k}_2|$. Also, in such a two-photon extension, nontrivial geometries are allowed, such as an oblique incidence for the irradiating beams, with $\vec{k}_1+\vec{k}_2$ remaining along the normal. This versatility may be used as an advantage by choosing an irradiation close to the Brewster angle (provided that the wavelengths of pump and probe are not too different) (see Fig. 2, and [9]), hence eliminating the Fabry-Perot effects [7] associated with extremely thin cell (ETC) spectroscopy. Also, it opens the possibility to optimize, with the choice of incidence angles, the Dicke narrowing for a given cell thickness.

The above description allows for a comparison between the co- and counterpropagating geometries. When the two irradiating frequencies are nearly equal ($\omega_1\approx\omega_2$), a counterpropagating geometry provides a negligible Doppler broadening in a macroscopic cell, while the copropagating geometry is affected by a Doppler broadening twice as large as the one for one-photon transition. In contrast, for an ETC with a thickness suitable for the two-photon Dicke narrowing

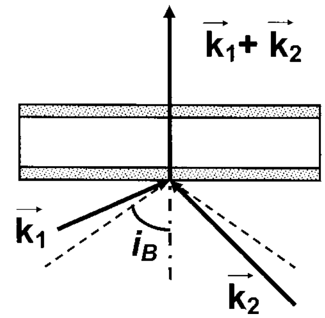

FIG. 2. A noncollinear geometry, close to the Brewster angle i_B, enabling us to observe the two-photon Dicke narrowing when $(\vec{k}_1+\vec{k}_2)$ is perpendicular to the thin cell.

in the copropagating geometry, i.e., $L=\Lambda/2=\lambda_1\lambda_2/2(\lambda_1+\lambda_2)(\approx\lambda_1/4\approx\lambda_2/4)$, the counterpropagating geometry corresponds to an ETC thickness much smaller than the optimal one (i.e., $L\ll\Lambda$ with $\Lambda=\lambda_1\lambda_2/|\lambda_1-\lambda_2|$), implying a spectral regime dominated by transit time effects, so that the two geometries provide comparable spectra (see Fig. 3). Moreover, when ω_1 and ω_2 largely differ, the sub-Doppler Dicke narrowing can still be attained (in the copropagating geometry for a specific well-chosen thickness, or if preferable, with an adapted oblique geometry). Note that the counterpropagating geometry, that would be sensitive to a partial Doppler broadening in a macroscopic cell, actually undergoes a reduction of this Doppler broadening through transit time effects, at the expense of a drastic reduction of the contributing velocities. Besides, a frequency-modulation (FM) technique [3] can turn these narrow spectra into Doppler-free lineshapes.

The possible observation, and benefits, of such a two-photon coherent narrowing could be limited by the large one-photon detuning Δ, with respect to a signal decreasing with $1/\Delta^2$. Because tunable sources open the possibility to strongly increase the strength of a two-photon signal by coming close to a nearly resonant one-photon excitation, it is worth discussing practical limitations. It is essential for the validity of (3) that Δ (or D_{21} and D_{32}, see the appendix) largely exceeds $|v_z|/L$. In spite of the fact that the two-photon Dicke narrowing (for $L=(2n+1)\Lambda/2$) induces a sub-Doppler structure through an overweighed contribution of

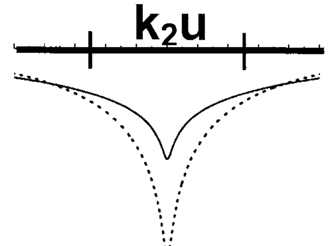

FIG. 3. A comparison between the copropagating geometry (full line), and the counterpropagating geometry (dashed line). The conditions are $\omega_{12}=\omega_{23}$, $\lambda_1\approx\lambda_2$, $L=\lambda_1/4=\lambda_2/4$, enabling strict Doppler-free two-photon spectroscopy in a macroscopic cell. One has taken $\gamma_{31}=k_2u/40$. The calculation is independent of the detuning Δ.

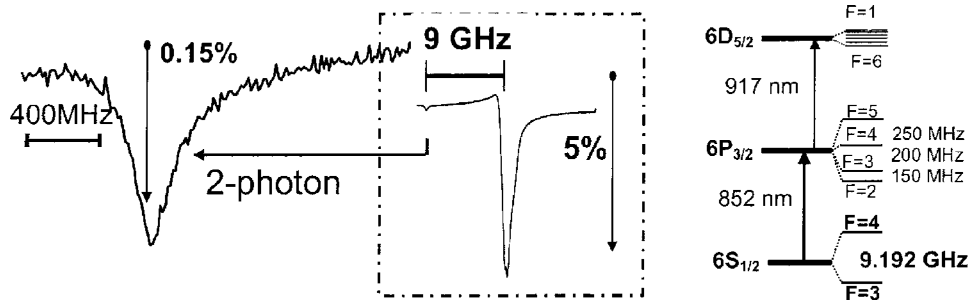

FIG. 4. Transmission spectrum recorded across the excited transition $6P_{3/2}-6D_{5/2}$ (917 nm) in a counterpropagating geometry on a 315-nm-thick Cs cell ($T=220$ °C) with the pump beam locked onto the $6S_{1/2}(F=3)-6P_{3/2}$ transition. The spectrum on the left is a zoom on the auxiliary resonance corresponding to the two-photon resonance $6S_{1/2}(F=4)-6D_{5/2}$. The pump frequency is locked to the $6S_{1/2}(F=3)$ $-6P_{3/2}$ transition. The hyperfine structure of the $6S_{1/2}-6P_{3/2}-6D_{5/2}$ levels is also shown.

the slow atoms, the coherent (and constructive) contribution of atoms with thermal velocities cannot be neglected, even at the line center, and the requirement exp-$[\Delta\Lambda/2v_z]\ll 1$ applies also to fast (i.e., thermal) atoms, practically implying for the detuning to be in excess of several Doppler widths. However, for an oblique and large angle incidence (e.g., Fig. 2), the two-photon Dicke-type narrowing could remain observable, even for a relatively weak detuning Δ. Indeed, the successive one-photon processes cannot eliminate the Doppler broadening when two different $\vec{k}_1$ and $\vec{k}_2$ axes are involved, so that the stepwise excitation process cannot strongly interfere with the two-photon process. As a counterpart of the large pump detuning, the pump intensity can be high, as the condition Ω_1, $\Omega_2\ll|\Delta|$ required by the third-order perturbation expansion, that imposes a Stark shift much smaller than Δ, remains easily respected.

Finally, this is because of these sensitivity issues that we report below our observation of a direct two-photon transition in a submicrometric cell, obtained through an extended frequency scan as a side effect in an experiment [10] actually devoted to ETC spectroscopy on the stepwise $6S_{1/2}$ $-(6P_{3/2})-6D_{5/2}$ transition of Cs. Indeed, although the experiment was performed in a counterpropagating geometry not enabling the specific observation of the Dicke narrowing, it yields instructive information about the signal magnitude. The pump frequency ω_1, locked onto the one-photon 852-nm resonance for the $6S_{1/2}(F=3)\rightarrow 6P_{3/2}$ transition, is indeed a blue-detuned pump for the $6S_{1/2}(F=4)\rightarrow 6P_{3/2}$ transition, with the $\Delta=9.192$ GHz detuning largely exceeding any relevant width for a two-photon transition. This justifies that when scanning the 917-nm probe on the $6P_{3/2}-6D_{5/2}$ transition, one observes (Fig. 4), aside from the stepwise two-photon resonance, a redshifted resonance associated to the direct two-photon transition $6S_{1/2}(F=4)\rightarrow 6D_{5/2}$.

The main information derived from this recording is that, in spite of the unusually small size ($L=315$ nm) of the cell, the two-photon signal is observable, and here of a relatively large size. The absorption is in excess of 10^{-3}, and the signal-to-noise ratio is easily improvable, as for the specific purpose of the stepwise excitation experiment, the sensitivity of the setup was nonoptimized and limited to $\sim 10^{-4}$. Also, the two-photon signal, observed here for a moderate pump intensity (1 mW focused on a ~ 100 μm spot, i.e., $\Omega_1\sim 250$ MHz), could be largely enhanced by decreasing the 9.192-GHz pump detuning, while remaining in the frame of a nonresonant two-photon transition. These potential improvements should enable the observation of the two-photon signal at a lower atomic density, the effect of which seems to be the dominant reason for the unexpectedly broad two-photon signal (the $6D_{5/2}$ hyperfine structure accounts only for ~ 50 MHz, the counterpropagating geometry only allows an ~ 20-MHz two-photon Doppler broadening, and the transit time is typically in excess of 1.5 ns, i.e., a broadening below 100 MHz). It can be also noted that the direct two-photon signal is less sensitive to the pressure broadening than the stepwise excitation because resonance collision processes affect the intermediate transitions.

In conclusion, we have demonstrated that the recently demonstrated coherent Dicke narrowing in an ETC can be extended through versatile setups to two-photon transitions, and a genuine experimental observation appears quite feasible. Even if the Dicke narrowing may remain only of a marginal interest for precision spectroscopy (the fabrication of ETCs [4,11] requires thick windows for a small active thickness), several possible applications can be envisioned. ETCs are already an interesting tool for the probing of atom-surface interaction, notably for the effective analysis of the spatial dependence of long-distance atom-surface interaction [10]. The high-lying excited states, of a special interest in this context, can be conveniently reached with two- or multiphoton excitation. Direct transitions from the ground states are naturally simpler to analyze than multiple stepwise excitation; in particular there is no need to assume a quasithermal pumping (see e.g., [12]), hardly justified in an ETC, and that ignores the surface interaction that shifts the one-photon resonance. Moreover, because in an ETC there is an upper limit to the atom-surface distance, the effect of atom-surface interaction in a multiphoton transition should be simpler to analyze in ETC spectroscopy than in the alternate selective reflection (SR) spectroscopy (see [12]). In this last case, the effective length of the probed region under multiple beam irradiation has never been determined unambiguously in spite of various principle analysis [13]. Extension of the multiphoton Dicke narrowing to a Λ-type systems based upon Raman transitions in a counterpropagating geometry can also be predicted, with $\vec{k}_1-\vec{k}_2$ the relevant wave vector. For Raman transitions between sublevels of the ground-state, the expected long lifetime of the Raman coherence should make observable a large number of collapses and revivals of the Dicke narrowing (for Cs, and a ground-state coherence lifetime assumed to be $\sim 1\ \mu s$, the decoherence length is $\sim 200\ \mu m$, to be compared with a Dicke periodicity $\Lambda = 426$ nm for an excitation close to the D_2 line with counterpropagating beams). Hence, with relatively long cells (~ 10–$100\ \mu m$), a different type of ground-state Raman coherence analysis appears feasible. The effective decay of the Raman coherence narrowing, measured through an adjustable cell length or through an angular adjustment of the respective exciting beams, could yield sensitive information on how the environment—and confinement—affects the details of the relaxation of the Raman coherence.

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APPENDIX

We give below the spatial dependence of the transient optical coherence σ_{32}. The calculation is obtained in the frame of a third-order perturbation expansion, and for the sake of simplicity, a conservative atomic system is assumed, with negligible dephasing collisions. In spite of the nonresonant nature of the pump excitation, the rotating wave approximation is performed also with respect to the pump, simply assuming that $|\omega_1 - \omega_{12}| \ll |\omega_1 + \omega_{12}|$. Our scope is indeed to define conditions for which the elementary two-photon processes dominates in the competition with the nearly resonant stepwise (one-photon) processes. Under such hypotheses, one gets (for $v_z > 0$)

$$\sigma_{32}(z,v) = \frac{i\Omega_1^2\Omega_2}{8}\left[\frac{[1-\exp-(D_{31}z/v)]}{D_{31}(D_{21}-D_{31})(D_{32}-D_{31})} + \frac{[1-\exp-(D_{21}z/v)]}{D_{21}(D_{21}-D_{32})(D_{21}-D_{31})} + \frac{[1-\exp-(D_{32}z/v)]}{D_{32}(D_{32}-D_{21})(D_{32}-D_{31})} + \frac{[1-\exp-(D_{31}z/v)]}{D_{31}|D_{21}|^2}\exp(-D_{21}^{*}z/v) + \frac{[1-\exp-(D_{32}z/v)]}{D_{32}|D_{21}|^2}[1-\exp(-2\gamma_{21}z/v)]\right], \tag{A1}$$

with $D_{32} = \gamma_{32} + i\vec{k}_2\vec{v} - i\Delta_2$; $\gamma_2 = 2\gamma_{21}$, $\gamma_{31} = \gamma_3/2$, and $\gamma_{32} = (\gamma_3 + \gamma_2)/2 = \gamma_{31} + \gamma_{21}$. Note that because of the absence of dephasing collisions, one has $D_{31} = D_{32} - D_{21}^{*}$. In (A1), all terms but two exhibit a denominator on the order of Δ^3 (assuming that the one-photon detuning Δ largely dominates over all other broadenings). Among the two terms with a resonant denominator on the order of $D_{31}\Delta^2$, the first one is simply the two-photon coherence contribution appearing in (3) (after some further simplifications). Surprisingly enough, the other one originates in a transient buildup of population in the relay level $|2\rangle$, but the exponential factor shows that the contribution associated with the buildup of population induced in the level $|2\rangle$ is limited to a very short duration $\sim 1/\Delta_2 (\approx 1/\Delta)$, during which the system does not have time to experience the one-photon detuning. Hence, for any reasonable length of the ETC, only the resonant two-photon coherence survives.

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