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Collapse and revival of a Dicke-type coherent narrowing in a sub-micron thick vapor cell transmission spectroscopy

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PACS. 42.50.Gy – Effects of atomic coherence on propagation, absorption, and amplification of light; electromagnetically induced transparency and absorption.
PACS. 32.70.Jz – Line shapes, widths, and shifts.
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Abstract. – In a thin cell of dilute vapour, the absorption spectrum exhibits sub-Doppler features due to the relative enhancement of the slow atom contribution, with respect to the transient nature of the interaction with moving atoms. For a two-level system in the linear regime, the narrowest response is predicted to be found for a $\lambda/2$ thickness, as an effect of the coherent character of the dipole response as early described by Romer and Dicke (*Phys. Rev.*, **99** (1955) 532) in the microwave regime. We report here on the direct observation of this effect in the optical regime in an ultra-thin vapour cell. This effect is shown to vanish for a thickness equal to λ, and a revival is observed at $3\lambda/2$, as expected from the predicted λ-periodicity. The experiment is performed on the D_1 resonance line of Cs vapour ($\lambda = 894$ nm), in a specially designed cell, whose thickness varies locally.

The resonant absorption of a very thin cell, filled with a dilute vapour, does not feature a simple Doppler-broadened lineshape when irradiated by light under normal incidence: because the atoms fly wall-to-wall, where they return to the ground state, the duration of the atom-light interaction is anisotropic. The contribution of atoms with slow normal velocity is enhanced thanks to their longer interaction time with the e.m. field. This principle has yielded novel possibilities for sub-Doppler laser spectroscopy [1–5]. A series of observations has relied on the relatively slow process of optical pumping in multilevel systems [1, 2, 4], and was performed with (relatively) long cells (thickness between 10 μm and 1 mm). The optical pumping process, enhanced for slow atoms, *decreases* the light absorption on line center [1, 4].

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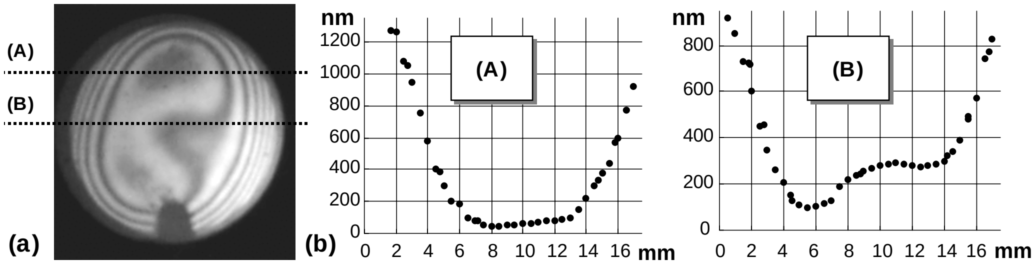

Fig. 1 – The extremely thin Cs vapour cell: (a) photograph of the cell, under a backward broadband illumination; (b) mapping of the cell thickness, along the respective lines (A) and (B) as drawn on the photograph.

The extension to *coherent* dipole absorption in a two-level system has appeared to be more experimentally demanding. The *increased* linear absorption of slow atoms has been monitored on the transmission of 10–50 μm cells under ultra-weak irradiation ($< 1\,\mu\mathrm{W/cm}^2$). This observation has been made possible thanks to the enhanced spectral sensitivity of a frequency modulation technique [3, 4]. However, the coherent character of the transient dipole has not yet been observed.

The purpose of this letter is to report on the coherent properties and oscillatory behaviour of light transmission of ultra-thin gas cells, the thickness of which, L, is on the order of the light wavelength, λ. The cell absorption is thus governed by the transient response of the induced electric-dipole moment [3]. Because of the sudden excitation triggered when an atom leaves one of the cell walls in its ground state, the induced optical dipole precesses in phase with the e.m. field at the wall position. A dephasing factor relative to the incident field starts to build up as the atom flies between the two walls, reaching the final value $\exp[ikL]$ at the other wall ($k = 2\pi/\lambda$). This phase mismatch $\Delta\varphi = kL$, analogous to the one observed in nonlinear optical generation, is proportional to the cell thickness, L, introducing a coherence length $\pi/k = \lambda/2$ in the interaction process. It subsequently modulates the coherent dipole emission with a period equal to λ: the sub-Doppler coherent emission, integrated over the cell length L, is hence predicted to reach a maximum value for $L = \lambda/2$, vanish for $L = \lambda$, and reappear at $L = 3\lambda/2$ [3, 6–8].

Pioneering work has been performed long ago in the microwave domain by Romer and Dicke [9] on the ammonia transition ($\nu = 24\,\mathrm{GHz}$). Using a standing-wave excitation, they observed a spectral response, narrower than the inhomogeneous width, in a "pillbox" cell of thickness $\lambda/2 = 1.25\,\mathrm{cm}$. In the optical domain, the observation of the coherent nature of the transient dipole response is made more difficult because of the smaller wavelength, and because, due to the coupling with vacuum fluctuations, the finite lifetime τ of the optical dipole introduces a decoherence effect in the oscillatory behaviour of the cell transmission: as soon as the cell length becomes larger than the dipole mean free path $\langle l \rangle = u\tau$ (u: mean atomic velocity, $u = (2k_\mathrm{B}T/M)^{1/2}$, with T the temperature, and M the atomic mass), the spatial oscillations of the sub-Doppler transmission spectrum are washed out. Previous observations of sub-Doppler coherent spectra [3, 4] have been performed in a range of cell thickness ($L \geq 12\lambda$) such that the predicted oscillatory behaviour could not be observed because of decoherence and also because of cell imperfections.

Recently, a series of Extremely Thin Cells (ETCs) of alkali-metal vapour, with $100\,\mathrm{nm} < L < 1\,\mu\mathrm{m}$, could be prepared [10], and narrow sub-Doppler absorption and fluorescence spectra have been directly observed with such ETCs. A description of the fabrication method of these cells has been given in [10]. In spite of the strong thickness and extreme flatness of the YAG windows that are used for the cell fabrication (these windows are contacted thanks to an evaporated spacer of aluminium oxide, then glued with a special gluing technology), the evacuated cell, sensitive to the external atmospheric pressure, exhibits notable local thickness variations: in a large region (see fig. 1a), the internal reflection in the visible range is very low, indicating a nearly close contact between the two windows, while on the edges, a few circular fringes are observed. These local variations, although a key for our thickness-dependence analysis, are distributed among such a large area that they confirm the excellent parallelism of the windows (largely better than 0.1 mrad). For this reason, the cell has to be considered as a Fabry-Pérot (FP) system, and this has two consequences: i) it induces an intrinsic mixing of reflection and transmission behaviours, that has to be taken into account according to the analysis made in ref. [8]; ii) non-resonant impinging light follows a nearly ideal FP behaviour, whose reflectivity is provided by the Fresnel formulae (index of a YAG window: $n = 1.80$), allowing an accurate evaluation of the local cell thickness. Following a first mapping of the cell (fig. 1b), an accurate technique of *in situ* thickness measurement (currently at the ± 4 nm accuracy level) was developed with a simultaneous 3-wavelength measurement ($\lambda_1 = 633$ nm, $\lambda_2 = 894$ nm, $\lambda_3 = 1298$ nm) [11]. Although a heating of the Cs vapour (in the 100–150 °C range) was required to get a sufficient vapour density and number of interacting atoms, we have verified that the local cell thickness —measured at the actual temperature of operation— was insensitive to this heating [12].

A systematic study of the transmission and reflection spectra was conducted as a function of the cell thickness L, in the regime of linear atom-field interaction. The laser is a DBR-type diode laser, whose spectral linewidth, on the order of 3 MHz, is narrower than the width of the Cs D_1 transition (natural width: 5 MHz). In some series of experiments, the laser diode was frequency modulated (typically at a frequency $f = 12$ kHz, and with an amplitude of $\Delta = 10$ MHz) by modulating the injected current ($\Delta I = 13\,\mu$A). The typical spot diameter was limited to $\sim 200\,\mu$m, to optimise the accuracy on the cell thickness. The laser-irradiating power was kept at a low value (~ 100 nW, *i.e.* $\sim 0.3\,\text{mW/cm}^2$) to avoid saturation effects. The linear behaviour has been checked systematically. This has occasionally enabled us to increase the intensity for short thickness, when the resonant signal drops down to low values, while the limited interaction time repels the onset of saturation effects to a higher value. Even for the thicker regions, the overall maximal absorption has been limited to a few percents, in order for the driving field amplitude to remain constant along the propagation axis.

To perform a sensitive spectral analysis, low-noise photodetetectors were used, with an adjustable gain in order to maximize the level of the absorption signal, relatively to the background electronic noise. Transmission and reflection signals were monitored simultaneously. For those measurements that were performed directly (*i.e.* in the absence of a FM technique, that permits one to use a sensitive lock-in detection along with a frequency transfer of the signal at the modulation frequency), a differential technique was implemented, to subtract most of the laser power variation during the frequency scan. Moreover, the tuneable DBR laser seems to exhibit a negligible frequency drift, if any, when successive frequency scans are operated. This enabled us to integrate numerous successive quick frequency scans (typically, 128 spectra at several MHz/ms). Hence, the effect of the low-frequency noise (*e.g.*, d.c. noise in the injection current) is randomized over the whole spectrum, instead of generating arbitrary frequency distortions in the lineshape.

Figure 2 presents the transmission lineshapes across the $6S_{1/2}(F = 4) \rightarrow 6P_{1/2}(F' = 4)$ hyperfine component, as a function of the cell thickness L, with the experimental spectra (fig. 2a) compared to a theoretical modelling (fig. 2b) [8]. An analogous behaviour has been observed across the various hyperfine components of the D_1 line, and we discuss here the most

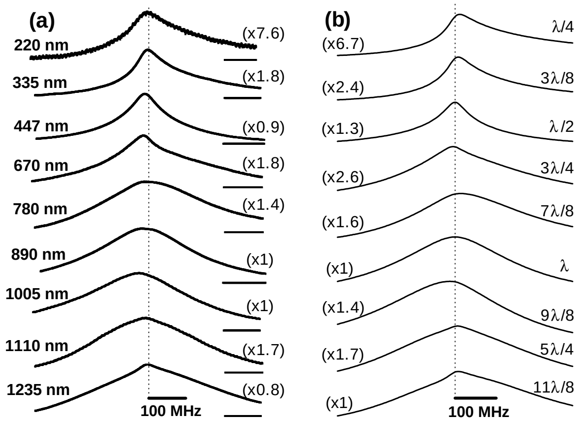

Fig. 2 – (a) The transmission spectrum observed on the $F = 4 \to F' = 4$ hyperfine component of the Cs D_1 line ($\lambda = 894\,\text{nm}$), in the Cs cell heated at $\sim 118\,^{\circ}\text{C}$, for local thickness as indicated. The dotted vertical line is a frequency marker, provided by an auxiliary saturated absorption experiment in a macroscopic cell; (b) theoretical spectrum for the corresponding thickness, assuming $\gamma/ku = 0.075$ The horizontal scale is identical to the one of the experiment in (a) when taking $ku = 250\,\text{MHz}$. For both sets of curves, a single (arbitrary) vertical unit is used, once the indicated sensitivity changes are considered.

remarkable results concerning the linewidth, symmetry, and amplitude of these spectra: i) As shown previously [10], the lineshape of the ETC transmission is narrower than the Doppler width. Actually, there are notable changes in the lineshape with the cell thickness, that have not been observed previously. Around $L = \lambda/2$, the lineshape exhibits a maximal narrow contribution. This narrow contribution vanishes when increasing L up to λ. One can also distinguish for $L \geq \lambda$ a revival of this narrow contribution, that appears superimposed to the broadened sub-Doppler structure. As illustrated in fig. 3, the FM transmission, with its intrinsic emphasis on the narrow structures, shows even more convincingly this collapse and revival of the narrow structure, that relates with the "coherent Dicke narrowing" mentioned above. Note that for this observed first revival, the maximum, predicted for $3\lambda/2$ (*i.e.* $1.34\,\mu\text{m}$), is not indeed attained in our experiments since $L \leq 1300\,\text{nm}$. Also, there is an imperfect vanishing for $L = \lambda$ that may partly relate to the local averaging implied when defining the cell thickness L (on both sides of $L = \lambda$, the narrow component re-appears with the same sign). ii) The lineshape exhibits a marked asymmetry for most values of the cell thickness L, a symmetric lineshape being observed only for $L \sim \lambda/2$ and $L \sim \lambda$. This is essentially a consequence of the FP behaviour of the ETC, so that —as discussed at length in ref. [8]— the expected symmetric absorption (transmission) signal mixes up with the back-reflection of the induced backward field. This (non–phase-matched) emission that is responsible for the selective reflection signal (*i.e.* it induces the resonant changes in the reflection coefficient) is indeed partially governed by a dispersive (antisymmetric) behaviour. This mixing is particularly visible in the wings of the resonance, when the dispersion dominates over the absorptive response. iii) The amplitude of the absorption evolves in a complicated manner: in spite of an overall growth of the amplitude with increasing L for a constant vapour temperature or density, the oscillating amplitude of the narrow contribution occasionally leads to a decrease —at line centre— of the absorption when L increases. Also, the FP-type behaviour affects the non-resonant transmission as well, and has an influence over the resonant transmission signal.

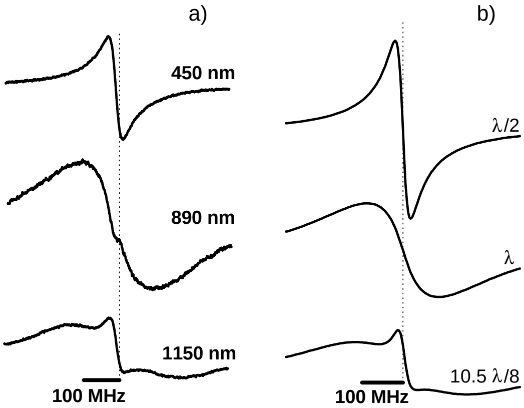

Fig. 3

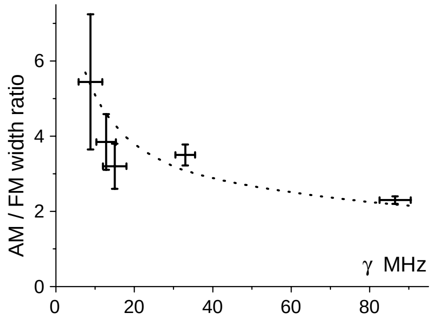

Fig. 4

Fig. 3 – The FM transmission spectra for different cell thicknesses, as observed with a lock-in detector: (a) experimental spectra for Cs D_1, $F = 4 \rightarrow F' = 4$; (b) theoretical predictions (ref. [8]). The FM frequency is 12 kHz, its p-p amplitude is $\sim$ 10 MHz. Note the change in the frequency scale relatively to fig. 2. Although the linewidth remains well below the full Doppler broadening for all the cell thicknesses considered here, the narrower contribution is clearly eliminated for the 890 nm ($\sim \lambda$) thickness.

Fig. 4 – A study of the line width of the transmission signal in the optimal situation of a $\lambda/2$ thick cell. The ratio of the (FWHM) width when comparing direct transmission to FM transmission is plotted as a function of the optical width γ, which is controlled through changes in the Cs vapour temperature. The dotted line is the theoretical prediction of the model (for $ku = 250$ MHz). Decreasing the Cs pressure induces a stronger linewidth narrowing in the FM technique than in the direct transmission technique. The width γ has been estimated with the theory (assuming $ku = 250$ MHz) from the measured FM width (FWHM). Note that the width of the FM signal is 2γ (FWHM) as long as $\gamma \ll ku$.

It even justifies the observation, in the resonance wings, of a net increase in the transmission, when L is close to multiple values of $\lambda/2$. Indeed, the optical length of the cell is modified by the dispersion that is dominant in the wings in such a way that the amount of (non-resonant) transmission can become closer to its peak value —the relative change of the optical length being currently in the 10^{-3} range.

In figs. 2b and 3b, the theoretical transmission lineshapes, plotted as a function of the cell thickness L, are evaluated in the framework of a two-level model [8]. With the single adjustable parameter chosen to be $\gamma/ku = 0.075$ (with γ the homogeneous linewidth, and ku the Doppler width), the agreement with the experimental lineshapes is striking, regarding both the details of the various lineshapes, the amplitude dependence on L, and even the slight frequency shift imposed to the peak of the transmission by the (L-dependent) residual asymmetry. Note that with the estimated value $ku \approx 250$ MHz (for a 120 °C Cs cell), it yields a homogeneous width $\gamma \sim 19$ MHz. Such a homogenous width is large relatively to the natural width of the Cs D_1 line (5.3 MHz), to the $\sim$ 3 MHz linewidth of the DBR laser, and to the FM broadening, and should be attributed to some pressure broadening. Indeed, because the signal level decays sharply for thickness below $\lambda/2$, and the series of measurements has been performed at small distances like $\lambda/4$ —and even, in experiments to be reported elsewhere, for smaller L values, for which the van der Waals surface interaction induces visible modifications [13]— the atomic

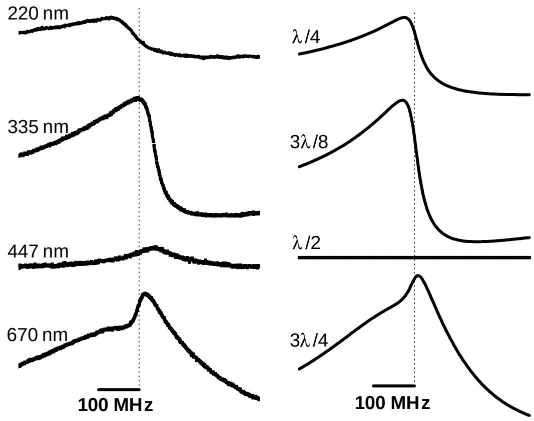

Fig. 5 – Comparison between experimental (left side) and theoretical reflection spectra (right side) for various cell thicknesses. The vertical dotted lines are a frequency marker set at the atomic transition frequency.

density is relatively large. At a thickness $L = \lambda/2$, when the linewidth is minimum, narrower lineshapes could be observed by decreasing the temperature —and hence γ/ku in the model— (see fig. 4). The linewidth (FWHM) has dropped from $\sim$ 150 MHz down to $\sim$ 80 MHz, and in the FM mode, from 40 MHz to $\sim$ 25 MHz. In a similar manner, when compared to an auxiliary saturated absorption spectrum, the ETC transmission lineshape is apparently red-shifted by an amount ($\sim$ 10 MHz) depending on the considered hyperfine component, also to be attributed to a collision effect (that will be analyzed elsewhere).

The above observations and discussions are confirmed with the simultaneous recording of reflection spectra, as illustrated in fig. 5 for various cell thicknesses. Practically, one notices that, in the mixing of effects associated to the coherent narrowing in ETC on the one hand, and to the FP structure of the ETC on the other hand, the latter effects induce dramatic effects in the reflection spectra, with the detection sensitivity rapidly oscillating with L. This relates with the quasi-nulling of the non-resonant FP reflection, with its recognizable $\lambda/2$ periodicity, as opposed to the λ periodicity of the revival of the coherent narrowing. Quick variations of the lineshape are also noticeable, from dispersion-like features —notably in the wings— to absorption-like ones at line centre. This is an effect of the multiple transmissions that go along with the reflection on the second window. As expected, there is a relative increase of the absorption-like features when L increases.

In conclusion, our experiments confirm that spectroscopy in an ETC is a very convenient way to get a sub-Doppler resolution; they also show a strong dependence on the cell thickness, with the $\lambda/2$ thickness an optimum in frequency resolution as well as in signal strength. It is the first experimental demonstration, moreover in the optical domain, of an early prediction [9] of coherent narrowing, that had been initially based upon qualitative arguments, apparently applicable also to a standing wave irradiation. It should be stressed that an incoherent model of spectroscopy in an ETC, as previously developed for optical pumping effects [1,2,4], cannot predict the observed revival of the narrow structure for $L \sim 3\lambda/2$, that the FM technique makes easily observable. Conversely, the model presently used [3, 6, 8] specifically involves the coupling between the phase of the absorption transient build-up, and the corresponding Doppler shift. More generally, this simultaneous optimisation of the absorption, and the sharp narrowing for $L = \lambda/2$, may offer an attractive tool, especially in its FM version, for the development of sub-Doppler molecular references, when optical saturation is difficult to reach. Extension of the present results to a stronger confinement like a 2D confinement (*e.g.*, a vapour confined inside some photonic crystal or photonic crystal fibers [14]) is also under consideration. At last, we have recently performed a series of ETC transmission experiments between excited states which establishes that ETC spectroscopy is very appropriate for the detection of long-range atom-surface interaction, including the possibility of a spatial resolution of the interaction. A precise analysis of the spectroscopic and FP behaviour of an ETC when the long-range surface vW attraction can be neglected, as is the case in the present paper (the vW interaction lies here [15] in the range of a few kHz μm^3, *i.e.* few MHz at 100 nm), is a natural prerequisite to a measurement of the atom-surface interactions in more complex situations.

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[11] The measured internal windows reflection follows a low-finesse Airy function, measured as a ratio with the reflection coefficient of the outer window. Scattering losses attenuate the expected peak value by less than 5%. Due to the 3-wavelength measurement, a high sensitivity is obtained because, for at least one wavelength, the slope of the Airy function exhibits a large value. Note that further experiments on an analogous sapphire cell have shown that the scattering losses can seriously impair the cell thickness evaluation.
[12] This is a consequence of the choice of using thick YAG windows. Further cell technology developments show that, with thinner windows, the local thickness can be temperature-controlled; it can also be made tuneable with a variable external pressure. Note also that, when increasing temperature, we observed an increase of scattering losses, probably due to some Cs layer.
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