Doppler-free spectroscopy of the lowest triplet states of helium using double optical resonance

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Abstract. Optical pumping on the 2s–2p transition (1083 nm) of metastable ³He or ⁴He atoms is used for fundamental science and applications. We report on its combination with an optical probe on the 2P–3S transition (706.5 nm) in a ladder-type double optical resonance scheme, with CW single-frequency pump and probe diode lasers. Theoretical expectations for level structure and absorption spectra were computed for both isotopes. Narrow Doppler-free absorption lines were experimentally obtained in low-pressure gas samples. The line weights and Zeeman structure in weak magnetic field agreed with expectations. The precision of the line positions and line splittings was limited by generic errors of our commercial Fizeau wavemeter, which were independently characterised. The potential of this double resonance scheme for spectroscopic measurements on the three lowest triplet states of He was evaluated. The relevance of velocity- and sublevel-selective pumping combined with polarisation spectroscopy of probe absorption for a study of pressure-dependent population transfers in the 2P state was established.

1 Introduction

The two lowest-lying triplet electronic states of helium are involved in the 1083 nm optical transition used to perform optical pumping (OP) of the 2S metastable state, which is usually populated by a radiofrequency (RF) discharge in a low-pressure gas. In ³He, OP is the driving term of metastability-exchange optical pumping (MEOP), an efficient process yielding high nuclear polarisations in the atomic ground state and used in many applications, ranging from fundamental physics to medical imaging [1]. Additional features resulting from polarisation in the 2S state are also investigated in gas discharges in ³He [2–4] or used in ⁴He for a variety of magnetometer schemes [5–7]. One important physical process affecting the operation and efficiency of the 2S–2P OP cycle is the redistribution of atoms among sublevels of the 2P state during the radiative lifetime as a result of collisions with ground-state atoms of the gas. Its pressure-dependent influence may be strong, yielding for instance a change in the sign of the OP-induced polarisation between low and high gas pressure [1, Sect. III.C.2].

In spite of the importance of 1S–2P collisions, detailed studies of collisional rates in RF gas discharges (as well as quantitative analysis of their contribution to OP dynamics for both isotopes) are still missing.

Only few theoretical evaluations [8,9] and one global indirect measurement [10] of the cross-sections for collisional transfer between ⁴He fine structure levels were reported. We therefore designed an experiment to measure collisional transfer rates within the 2P manifold for both isotopes and to establish correlations with OP dynamics features. Gaseous He samples were submitted to velocity-selective OP (VSOP) on the 2S–2P transition at 1083 nm and steady-state distributions of atomic populations and velocities in the 2P state were probed by laser absorption on the 2P–3S transition at 706.5 nm. A (co-propagating) collinear pump-probe configuration was chosen to maximise probe light absorption and to avoid residual Doppler broadenings at 706.5 nm (potentially large, compared to the natural widths, in He). Combination of absorption spectroscopy and polarimetry at 706.5 nm allowed quantitative assessment of atomic number densities in the probed 2P sublevels, as well as discrimination between atoms selectively excited by the pump laser and atoms transferred to other velocity classes or sublevels by collisions.

Initial evidence of population transfers due to 1S–2P collisions was very briefly reported for isotopic mixtures [11]. The results of a detailed study of collisional transfer rates between fine or hyperfine 2P sublevels will be reported in a forthcoming article. Here, we mainly focus on the 2S–2P–3S three-level ladder system involved in the measurements and on the

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analysis of double optical resonance signals obtained when the effect of collisions is weak. We report on expected and measured Doppler-free spectroscopic data for the $^2\text{P} \rightarrow ^3\text{S}$ transition at 706.5 nm in $^3\text{He}$ and $^4\text{He}$. We also provide an example of 706.5 nm absorption spectrum obtained by double optical resonance when strong collisional mixing occurs. The advantageous boost in signal amplitude associated with the use of 1083 nm pumping may be appreciated by comparison with prior Doppler-limited spectroscopic investigations at 706.5 nm in He gas cells and RF glow discharges, with techniques such as analysis of fluorescence at 706.5 nm in He gas cells and RF glow discharges, with techniques such as analysis of fluorescence spectra (yielding hyperfine splittings of $^3\text{He}$ levels) and isotope shifts [12,13], or pressure broadening [14] and pressure shift [15] in $^4\text{He}$ and optogalvanic detection of laser absorption at 706.5 nm [16] (yielding $^3\text{S}$ hyperfine splittings, with a data fitting accuracy that was comparable to that of Doppler-free saturated absorption measurements).

The present article is organised as follows. Section 2 describes the computed fine, hyperfine, and Zeeman components of the $^2\text{P} \rightarrow ^3\text{S}$ transition. Absorbance data at 706.5 nm and at 1083 nm are quantitatively linked with atomic number densities in the $^2\text{P}$ magnetic sublevels and spectral features of the double optical resonance signals are discussed. Section 3 describes the experimental setup and the main elements used for the measurements. The latter include a commercial wavemeter, whose behaviour and stability are characterised in Appendix A. Section 4 compiles experimental results obtained with pure or mixed helium isotopes, at various gas pressures in the millibar range. Typical double optical spectra are presented, together with recordings made at moderately higher pressure (up to 67 mbar) that illustrate the observed changes in line shapes. Zeeman-split spectra at 706.5 nm are compared with expectations at a few milliteslas and the influence of pump intensity on probe absorption signals is reported. Then, the potential of the method and of the experimental arrangement for Doppler-free spectroscopy is evaluated. Compiled low-pressure data sets are used for measurements of the hyperfine structure of the $^3\text{S}$ state and of the isotope shift of the $^2\text{P} \rightarrow ^3\text{S}$ transition. Finally, Sect. 5 provides a discussion of the current limits and assets of double optical resonance of metastable helium.

2 Theory

2.1 Atomic structures and optical transitions

The fine and hyperfine structures of the three lowest triplet states of helium involved in the $^2\text{S} \rightarrow ^2\text{P} \rightarrow ^3\text{S}$ double optical resonance scheme are schematically depicted in Fig. 1 for both helium isotopes. The structures of the $^2\text{S}$ and $^2\text{P}$ states give rise to the various energy shifts (listed in Table 1) for the sublevels of given $J$-values (for $^4\text{He}$) or $F$-values (for $^3\text{He}$) involved in the 1083 nm optical transition. The remaining degeneracies between magnetic sublevels are removed when a magnetic field is applied and the Zeeman effect becomes significant. The sublevel naming conventions depicted in Fig. 1, the magnetic-field-dependence of the sublevel energies, and the positions and weights of the optical transitions components can be found in Ref. [17]. The null-field positions are reproduced in Table 2 for convenience.

Using the same approach, the level structure of the $^3\text{S}$ state was computed in the decoupled representations ($L$, $S$ for $^4\text{He}$ or $L$, $S$, $I$ for $^3\text{He}$). It is indeed similar to that of the metastable $^2\text{S}$ state and differs mostly because of the $\sim3\%$ lower value of its hyperfine structure constant [18, Table 7]:

$$A_S (^2\text{S}) = 4493.13 \text{ MHz}, \quad A_S (^3\text{S}) = 4372.89 \text{ MHz}.$$ (1)
The upper part of Table 1 lists the null-field energy terms of the three $^3P_J$ levels of $^3\text{He}$. Their low-field linear variations are

$$E_p(4)(B) - E_p(4)(0) = g_J m_J \mu_B B + \mathcal{O}(B^2)$$  

with $g_J$ given in Table 1. The low-field linear variations for the $^3\text{S}$ and $^2\text{S}$ states are simply given by the linear Zeeman energy terms

$$E_p(4)(B) = g_S^* m_S \mu_B B,$$  

where $g_S^* \approx g_S = 2.00224 \ [19, \ Table 5]$. The effective Hamiltonian of Ref. [17] yields accurate energy differences only between sublevels of each of the three states considered here. Absolute optical transition energies cannot be inferred. However, energy or frequency differences between optical transitions can be computed from the level energies of Table 1. The results are displayed in Table 2 as a list of computed null-field frequency offsets (or frequencies, for compactness) $\xi / h$ (resp. $\tilde{\xi} / h$) from the highest- (resp. lowest-) energy component of the 706.5 nm (resp. 1083 nm) transition, for both isotopes. The energy differences $\xi$ and $\tilde{\xi}$ are usually displayed with one index (that of the line component) for null-field situations or with two indices (those of the relevant sublevels) in applied fields.

For $^3\text{He}$ they are written as

$$\xi_{jk} = [E_S^* (A_k) - E_S^* (A_5)] - [E_p(B_j) - E_p(B_7)]$$  
$$\xi_{ij} = [E_p(B_i) - E_p(B_7)] - [E_S^* (A_i) - E_S^* (A_5)].$$  

The frequencies of $^3\text{C}_n$ and $^3\text{C}_n$ are exactly opposite when they involve the same $F = 1/2$ hyperfine level as $^3\text{C}_1$ and $^3\text{C}_1$, namely for $n = 2, 4, 8$ (see the table in Fig. 1).

For $^4\text{He}$ the null-field line splittings solely result from the fine-structure of the $^2\text{P}$ level. The line positions with respect to the 706.5 nm $^3\text{He}$ lines are computed using the average of the two published values of the positive isotope shift differences between level energies of $^3\text{He}$ and $^4\text{He},$

$$\delta_{\text{iso}} = \Delta E / h (^3\text{S}_1) - \Delta E / h (^2\text{P}_0) = 594.106 \ \text{MHz}$$
Table 3 Values of the Doppler width parameters (generically noted $\Delta$) in GHz, for $^3$He and $^4$He and for both optical transitions. They are computed for a temperature $T = 300$ K as $\Delta = \nu \sqrt{2k_B T/M c^2}$ ($\nu$ is the optical transition frequency, $k_B$ the Boltzmann constant, and $M$ the atomic mass). The Doppler FWHM values are $2\Delta/\ln 2$

|       | $^3$He | $^4$He |
|-------|--------|--------|
| 1083 nm | $\Delta_3 = 1.1875$ | $\Delta_4 = 1.0284$ |
| 706.5 nm | $\Delta_3 = 1.8203$ | $\Delta_4 = 1.5764$ |

Table 4 $^3$He transition matrix elements $T_{jk}$ between sublevels $B_j$ and $^*A_k$ for $B = 0$. The color indicates the light polarisation vector: red for $\sigma_+$, black for $\pi$, and blue for $\sigma_-$. It corresponds to the angular momentum change between the half-integer $m_F$-values ($m_F$ and $^*m_F$, not displayed in this table)

|       | $^*A_1$ | $^*A_2$ | $^*A_3$ | $^*A_4$ | $^*A_5$ | $^*A_6$ |
|-------|--------|--------|--------|--------|--------|--------|
| B1 | 1/3 |  |  |  |  |  |
| B2 | 2/15 | 1/5 |  |  |  |  |
| B3 | 1/30 | 1/5 | 1/10 |  |  |  |
| B4 | 1/10 | 1/5 | 1/10 | 1/30 |  |  |
| B5 |  | 1/5 | 2/15 |  |  |  |
| B6 |  |  | 1/3 |  |  |  |
| B7 | 1.19456 | 0.12977 | 0.12977 | 0.12977 | 0.12977 | 0.00891 |
| B8 | 0.12977 | 0.02163 | 0.17302 | 0.12977 | 0.12977 | 0.00594 |
| B9 |  | 0.17302 | 0.02163 | 0.12977 | 0.12977 | 0.00297 |
| B10 |  |  | 0.12977 | 0.12977 | 0.12977 | 0.00297 |
| B11 | 0.07296 | 0.04864 | 0.24323 | 0.07296 | 0.06247 | 0.12494 |
| B12 | 0.02432 | 0.04864 | 0.24323 | 0.07296 | 0.06247 | 0.12494 |
| B13 | 0.00535 | 0.00357 | 0.00357 | 0.00357 | 0.00357 | 0.00357 |
| B14 | 0.00357 | 0.0059 | 0.00475 | 0.0059 | 0.0059 | 0.00475 |
| B15 | 0.00475 | 0.0059 | 0.0059 | 0.0059 | 0.00475 | 0.00475 |
| B16 | 0.00357 | 0.0059 | 0.0059 | 0.0059 | 0.00357 | 0.00357 |
| B17 | 0.03123 | 0.06247 | 0.09370 | 0.09728 | 0.04864 | 0.09728 |
| B18 | 0.09370 | 0.06247 | 0.03123 | 0.04864 | 0.09728 | 0.09728 |

Note that the $\approx 0.6$ GHz isotope shift is smaller than Doppler widths at room temperature (listed in Table 3). This results in fully overlapping Doppler-broadened spectra for the 706.5 nm lines (Fig. 2c and [12,13]), a feature which contrasts with the 32 GHz isotope shift for the 1083 nm lines (Fig. 2d).

Transition matrix elements $T_{jk}$ ($e_{\lambda}$) for $^3$He between sublevels $B_j$ and $^*A_k$ and $T_{jk}^{(4)}$ ($e_{\lambda}$) for $^4$He between sublevels $Z_j$ and $^*Y_k$ are computed for the three light polarisation states $e_{\lambda}$ ($\sigma_+$, $\sigma_-$, and $\pi$). Their values at null field are listed in Tables 4 and 5 and displayed in Fig. 2a, b. The empty cells correspond to null values of $T_{jk}$ (forbidden dipolar electric transitions, $|\Delta F| > 1$ or $|\Delta m_F| > 1$ for $^3$He, $|\Delta m| > 1$ for $^4$He).

For both optical transitions the matrix elements correspond to branching ratios for radiative decay and obey similar sum rules. For instance, partial sums over the lower sublevels for each upper sublevel in the $3^3S$ state verify

$$\sum_{j,e_{\lambda}} T_{jk}^{(4)} (e_{\lambda}) = \sum_{j,e_{\lambda}} T_{jk} (e_{\lambda}) = 1.$$
Additionally, the $^4\text{He}$ matrix elements of the two transitions verify exact relations:

$$T_{jk}^{(4)}(\sigma_{\pm}) = T_{kj}^{(4)}(\sigma_{\pm})/3, \quad T_{jk}^{(4)}(\pi) = T_{kj}^{(4)}(\pi)/3,$$

(10)

where the opposite helicities of light result from the opposite roles (upper or lower level) of the $^3\text{S}$ and $^3\text{S}$ states in the optical transitions. The $^4\text{He}$ matrix elements verify similar relations only for $B = 0$, with small differences (for instance of order $10^{-3}$ at $B = 0.1$ T) resulting from the difference in hyperfine structure constant values (Eq. 1):

$$T_{jk}(\sigma_{\pm}) \approx T_{kj}(\sigma_{\pm})/3, \quad T_{jk}(\pi) \approx T_{kj}(\pi)/3.$$  

(11)

### 2.2 Probe transmittance

Explicit expressions for the transmittance $T$ of a weak probe beam through a helium gas sample can be written when several simplifying assumptions are made: the probe light has one of the three polarisation states $e_{\lambda}$, the probe beam has a small transverse size (across which the populations of all states are uniform), and the probe intensity is weak enough for stimulated emission to be negligible. In this article, the probe beam is tuned to the 706.5 nm transition but most results in this section apply for a weak probe on the 1083 nm transition as well. In addition to the hat-decorated notations introduced in Sect. 2.1, we therefore use complementary generic notations for the lower (L) and higher (H) states of a transition in an isotope, with degeneracies $g_L$ and $g_H$, sublevels $L_p$ and $H_q$, and populations $l_p$ in the lower state. Note that $l_p$ are true populations (with $\sum a_i = \sum y_i = 1$) only for the $^3\text{S}$ state. The (pseudo)populations $b_j$, $z_j$, $u_k$, and $y_k$ are convenient parameters to obtain the number densities of the corresponding $^3\text{S}$ state sublevels from $\tilde{n}$, as detailed in Table 6, where the local metastable number density $\tilde{n}$ stands for the isotope-specific notations $n_m$ or $n_m^{(4)}$ of Ref. [17].

The probe light intensity $I_{pr}$ decreases with $z$, the position along the light path, when propagating in the sample because of absorbed and scattered light

$$-\frac{dI_{pr}}{dz} = h\nu_{pr}\tilde{n} \sum_{p,q} l_p \int dv_z \phi_{lp}(v_z) \tilde{\gamma}_{pq}(v_z),$$

(12)

in which $\nu_{pr}$ is the central probe laser frequency, $\phi_{lp}$ is the normalised velocity distribution in sublevel $L_p$, and $\tilde{\gamma}_{pq}$ is the velocity-dependent optical transition rate from $L_p$ to $H_q$. For a normalised frequency distribution $F(\nu - \nu_{pr})$ of the probe light intensity, the transition rates may be written as

$$\tilde{\gamma}_{pq}(v_z) = \tilde{K} I_{pr} \tilde{T}_{pq}$$

$$\times \int dv \mathcal{F}(\nu - \nu_{pr}) L_W \left[ \nu - \frac{\tilde{\xi}_{pq}}{\hbar} \left( 1 - \frac{v_z}{c} \right) \right]$$

(13)

in which $\tilde{T}_{pq}$ are the transition matrix elements that represent $T_{pq}$ for pump light absorption and $T_{pq}^{(4)}$ for probe light absorption, and $L_W$ is a normalised Lorentzian distribution of full width at half maximum (FWHM) $W$:

$$L_W(\nu - \nu^*) = \frac{2/(\pi W)}{1 + [2(\nu - \nu^*)/W]^2}.$$  

(14)

For the transition rate of Eq. 13, $W = \tilde{F}^*/2\pi$ is linked with the pressure-broadened damping rate of the L–H transition (cf. Sect. 2.3). The coefficient $\tilde{K}$ can be conveniently derived by noticing that each integral over $v_z$ in the right-hand side sum of Eq. 12 is a convolution of normalised functions, and is thus a normalised function of the probe frequency. For an unpolarised gas with
equally populated sublevels \( \hat{n}_L = \hat{n}_L / g_L \), with \( \hat{n}_L \) the number density in the lower level, Eq. 12 can therefore be identified with the usual expression of absorption cross section \cite{21}\:

\[
\sigma_a(\nu_{pr}) = -\frac{1}{\hat{n}_L I_{pr}} \frac{dI_{pr}}{dz} (\nu_{pr}) = \gamma \frac{\lambda^2}{8\pi} \frac{g_H}{g_L} g(\nu_{pr})
\]

(15)

where \( \gamma \) is the total rate of spontaneous emission from the higher level, \( \lambda \) the transition wavelength, and \( g \) a normalised line shape function. Summing relations like Eq. 9 over all higher sublevels but retaining only one of the three light polarisations yields \( \sum_{p,q} \hat{T}_{pq}(\epsilon_\lambda) = g_H/3 \), and the value of \( \bar{K} \) is derived

\[
h\nu_{pr} \bar{K} = \frac{3\lambda^2\gamma}{8\pi},
\]

(16)

This can equivalently be expressed in terms of the absorption oscillator strength \( f \) \cite{21}:

\[
h\nu_{pr} \bar{K} = \frac{3g_H h\alpha f}{g_H 2m_e},
\]

(17)

where \( \alpha \) is the fine-structure constant and \( m_e \) the electron mass. The ratio \( 3g_H / g_H \) is equal to 1 for the 1083 nm transition and to 9 for the 706.5 nm transition.

These relations can be further simplified in several situations. For instance, when the sample is optically thin or when velocity distributions are sufficiently broad, absorption only affects the probe intensity \( I_{pr}(z) \), not its frequency distribution \( F \). In particular, if \( F = L_\delta \) is a Lorentzian intensity distribution of width \( \delta \), the convolution in Eq. 13 simply yields a Lorentzian of width \( W + \delta \):

\[
\bar{\gamma}_{pq}(\nu_z) = \bar{K} I_{pr}(z) \hat{T}_{pq} L_{W+\delta} \left[ \nu_{pr} - \frac{\varepsilon_{pq}}{h} \left( 1 - \frac{v_z}{c} \right) \right].
\]

(18)

The probe transmittance is finally derived by integration on \( z \) over the cell length, \( L_{cell} \), of the right-hand side of Eq. 12 divided by \( \hat{I}_{pr} \). This simply introduces spatial averages \( \langle \hat{n}_p \rangle \) of the probed sublevel atomic densities (or the spatial average \( \langle \hat{n} \rangle \) if the populations are uniform, which is assumed in the following), and a polarisation-dependent spectral function noted \( \hat{S} \):

\[
-\ln \bar{T} = L_{cell} \langle \hat{n} \rangle \frac{3\lambda^2\gamma}{8\pi} S(\nu_{pr}), \text{ with }
\]

\[
\hat{S}(\nu) = \sum_{p,q} I_p \hat{T}_{pq} \int dv_z \phi_{pq}(v_z) L_{W+\delta} \left[ \nu - \frac{\varepsilon_{pq}}{h} \left( 1 - \frac{v_z}{c} \right) \right].
\]

(19)

For thermal velocity distributions in unpolarised gas, which occur in a weak discharge in the absence of pump beam, the thermal spectral functions \( \hat{S}_{th} \) are linear combinations of Voigt profiles. For low gas pressure and narrow-band probe light \( (W + \delta \ll \Delta, \text{ with values of Doppler width listed in Table 3}) \), \( \hat{S}_{th} \) approximately combines Gaussian profiles:

\[
\hat{S}_{th}(\nu) = \frac{1}{\Delta_{\sqrt{\pi}} \pi} \sum_{p,q} L_p \hat{T}_{pq} \exp \left[ -\frac{(\nu - \varepsilon_{pq}/h)^2}{\Delta^2} \right].
\]

(21)

The scaled dimensionless quantities \( \Delta_{\sqrt{\pi}} \hat{S}_{th} \) are plotted in Fig. 2c, d. For the 1083 nm transition this quantitative link between transmittance and number densities of absorbers in the relevant sublevels is indeed identical to that previously published \cite{17,22,24}. It applies to the 706.5 nm transition as well when replacing the generic parameters in Eqs. 19 and 21 with the specific ones.

When velocity distributions \( \phi_{pq}(v_z) \) depart from thermal equilibrium, \( \bar{S} \) retains the property that each integral over \( v_z \) is a normalised function of frequency, which means for instance that integration of resolved components of transmittance data can yield quantitative information on number densities in the corresponding sublevel. For instance, when the \( ^1D_0 \) line component is scanned in \(^4\)He using any probe polarisation, the area \( A_0^{(4)} \) of the absolute transmittance data is

\[
A_0^{(4)} = -\int_{\nu_{D_0}} dv_{pr} \ln \bar{T}(\nu_{pr})
\]

\[
= L_{cell} \langle n_m^{(4)} \rangle \frac{3\lambda^2\gamma}{8\pi} \hat{T}_{9k}^{(4)} (z_6 + z_8),
\]

(22)

where the index \( k \) depends on the choice of polarisation but all \( \hat{T}_{9k}^{(4)} \) are equal to 1/9 (see Table 5). For other lines, similar simple relations can be explicitly derived in particular for a \( \pi \)-polarised probe, a configuration experimentally studied in this work. When the \( ^1D_1 \) component is used, the populations \( z_6 \) and \( z_8 \) are probed with equal weights and

\[
A_1^{(4)}(\pi) = L_{cell} \langle n_m^{(4)} \rangle \frac{3\lambda^2\gamma}{8\pi} \hat{T}_{61}^{(4)} (z_6 + z_8).
\]

(23)

Similarly when the \( ^1C_n \) line components probe absorption by \(^4\)He atoms in the \( 2^3P_0 \) level \((n = 8 \text{ or } 9)\)

\[
A_n^{(\pi)} = L_{cell} \langle n_m^{(4)} \rangle \frac{3\lambda^2\gamma}{8\pi} \hat{T}_{18,k} (b_{18} + b_{17}),
\]

(24)

where \( k = 5 \) for \( n = 8 \) and \( k = 2 \) for \( n = 9 \).

These expressions relate probe absorption data to number densities of selected sublevels in the \( 2^3P_0 \) state. Besides, the total number density in the \( 2^3P_0 \) state can independently be linked with the absorbed pump intensity \( \Delta I_P \) using simple detailed balance considerations.

\[1\] The difference between Eq. 15 and the expressions used in Ref. \cite{21} results from the fact that optical angular frequency \( \omega \) is consistently used in that work, contrary to e.g., Ref \cite{22} and this article in which the optical frequency \( \nu \) is used. Reference \cite{21} was revised and some corrections were made in \cite{23} but Eq. 15 remained unchanged.
involving the $2^3P$ decay rate and the wavelength of the pumping transition:

$$\Delta I_P = L_{\text{cell}} \left[ \langle n_m \rangle \sum_{j=1}^{18} b_j + \langle n_m^{(4)} \rangle \sum_{j=1}^{9} \tilde{z}_j \right] \gamma hc/\lambda. \quad (25)$$

Discharge-dependent parameters are thus eliminated when considering the ratios of any of the line areas $\hat{A}$ (Eqs. 22–24) to the absorbed pump intensity (Eq. 25), which yields

$$\frac{\hat{A}/\hat{T}_{jk}}{\Delta I_P} = \frac{3^2 \gamma^2 \gamma \lambda}{8 \pi \gamma hc} \hat{R}, \quad (26)$$

where $\hat{R}$ is the fraction of the atoms in the $2^3P$ state which are in the probed sublevel(s), e.g., $R_0 = z_0/\sum_j z_j$ for a probe frequency scanning the $^3D_0$ line in pure $^4$He.

When the pump intensity is weak enough to induce negligible atomic alignment or orientation (population differences) in the $2^3S$ state and small enough populations in the $2^3P$ state (therefore negligible stimulated emission), an exponential intensity decrease results from light absorption in the gas. In that limit, the absorbed pump intensity linearly scales with the average pump intensity as

$$\Delta I_P^{(\text{lin})} = - \ln T_0 \langle I_P \rangle \quad (28)$$

where $T_0$ is the null-intensity limit transmittance of the pump beam. When Eq. 28 is substituted in Eq. 26, a linear increase of the areas $\hat{A}$ with the average pump intensity is obtained in the weak-pumping limit. It involves the factor in Eq. 27 and the fraction $\hat{R}$ which is of the order of 1 at low enough pressure:

$$\hat{A}^{(\text{lin})} = - \ln T_0 \hat{T}_{jk} \hat{R} \frac{3^2 \gamma^2 \gamma \lambda}{8 \pi \gamma hc} \langle I_P \rangle. \quad (29)$$

This sets the scale for the expected optical signals and is used for comparisons with experimental data.

### 2.3 VSOP and probe absorption linewidth

Velocity-selective optical pumping (VSOP, [25]) is the key first step of double optical resonance. With a narrow laser tuned close to an atomic resonance, only atoms with velocity projections close to zero are excited with a significant rate from the $2^3S$ to the $2^3P$ state. Velocity distribution profiles in these two states may therefore strongly differ from thermal distributions. This is illustrated in Fig. 3b, c for the simple case of isolated pump and probe transitions in $^4$He, namely with $\pi$-polarised pump light on the $D_0$ component (Fig. 3a).

The steady-state result of VSOP in helium can be described by the (position-dependent) populations and the associated normalised velocity distributions of the $2^3S$ and $2^3P$ sublevels, using the notations listed in Table 6. For the sublevels $Y_2$ and $Z_0$ directly addressed by the pumping light in Fig. 3a, typical velocity distributions are displayed in panels b and c, respectively. A narrow hole is burnt in the thermal velocity profile of the $Y_2$ level (a narrow peak correspondingly appears on top of thermal velocity profiles of levels $Y_1$ and $Y_3$, not shown), while populations are modified ($a_2 < a_1, a_3$). In contrast, the velocity distribution in the $2^3P$ state mostly results from the VSOP process, with a negligible thermal contribution from the radiative cascade in a weak discharge: it is characterised by a narrow peak with a negligible broad pedestal. These usual features of VSOP are directly relevant for the Doppler-free spectroscopy performed in this work, since the linewidth $\Delta \nu$ of 706.5 nm light absorption signals (Fig. 3d) partly results from the velocity distribution width $\Delta v_z$ of atoms in the probed $2^3P_0$ level.

If the pump intensity $I_P$ is low enough, $\Delta v_z$ solely results from the combination of the natural linewidth of the 1083 nm transition, of its pressure broadening, and of the pump laser spectral width $\delta$. If the latter is assumed to have a Lorentzian profile, all contributions simply add up to yield the FWHM of the velocity profile

$$\Delta v_z.$$
for $I_p = 0$

$$\Delta v_z |_{I_p=0} = \lambda [\gamma + 2w + 2\pi \delta]/2\pi. \quad (30)$$

In Eq. 30 the wavelength $\lambda$ of the pump converts frequencies to velocities, $\gamma$ is the radiative decay rate in the $2^3P$ state ($\gamma/2\pi = 1.626$ MHz [26, Table 14]) and $w$ is the pressure-dependent collision broadening rate ($w/\pi \approx 12$ to 18 MHz/mbar [9,24]). The line shape of the 706.5 nm absorption signal is a narrow Lorentzian profile which results (by convolution) from the velocity distribution profile in the pumped $2^3P$ level and the linewidth of the probed transition itself. Its total width is therefore the sum of two terms, here expressed in frequency units:

$$\Delta \nu |_{I_p=0} = \Delta v_z |_{I_p=0} / \lambda + I_{706}/2\pi. \quad (31)$$

The first part involves the width of the velocity distribution of atoms excited by the pump light obtained in Eq. 30, converted to frequency using the wavelength $\lambda$ of the probe. The second part, $I_{706}/2\pi$, combines widths arising from the radiative lifetimes of the $2^3P$ state ($\gamma/2\pi = 4.425$ MHz [26, Table 14]), collisional broadening of the 706 nm transition (33 MHz/mbar, [14]), and the instrumental linewidth of the probe laser. Altogether, the expected FWHM is the sum of contributions due to the radiative lifetimes (6.92 MHz), collisional broadening (52 to 61 MHz/mbar), and laser widths (typ. 7.5 MHz for pump and probe lasers having 3-MHz widths), adding up to $\Delta \nu |_{I_p=0} = 22$ MHz for 0.1 mbar, the lowest pressure used in our experiments.

At sufficient pump intensity, power broadening arising from saturation of the transition and from the creation of holes in velocity profiles is well documented for simple two-level systems [27]. Dedicated OP models are needed in the case of helium to establish a quantitative link between pump intensity and broadening of the velocity profiles in the $2^3S$ and $2^3P$ states. Such models usually involve a coarse phenomenological treatment of collisions and of relaxation [17,28,29] and their use to evaluate velocity profiles falls beyond the scope of this article. Let us only mention that pressure broadening of the velocity distribution is expected to retain its generic expression as a function of the pump intensity $I_p$:

$$\Delta v_z = \sqrt{1 + I_p/I_{sat}} \Delta v_z |_{I_p=0}, \quad (32)$$

where, however, $I_{sat}$ is an effective saturation intensity which depends on various parameters in the OP model.

### 2.4 Zeeman effects in double optical resonance

In this section, generic notations are used to correspond to $^3$He or $^4$He, but the higher and lower states are implied by the choice of the pump or probe transition. Considering a pump-induced transition from $L_i$ to $H_s$, with average pump frequency $\nu_p$ and level energy difference $\hbar \nu_{ij}$, the pump detuning $\nu_{ij} - \nu_p$ selects atoms in the $2^3S$ state which are suitably Doppler-shifted. Their average velocity $v_z^S$ and the corresponding velocity of the atoms in the $2^3P$ state are linked by

$$v_z^P - v_{recoil} = v_z^S = (\nu_{ij} - \nu_p) \lambda \quad (33)$$

where the recoil velocity $v_{recoil} = h/(\lambda \gamma)$ amounts to 12.3 and 9.2 cm/s for $^3$He and $^4$He, respectively ($\dot{M}$ is the atomic mass). For a $2^3P$-$3^3S$ transition frequency $\nu_{jk}$ from $L_j$ to $H_k$, the probe frequency $\nu_0$ needed for resonant absorption by atoms with the pump-selected velocity of Eq. 33 satisfies

$$\nu_{jk} - \nu_0 = \nu_z^P/\lambda \quad (34)$$

$$= (\nu_{ij} - \nu_p)/\lambda + h/(\dot{M} \lambda \lambda).$$

The Zeeman shift of the resonant probe absorption frequency $\nu_0$ in an applied field $B$ is obtained as the difference of values obtained using Eq. 34 for $B$ and a null field. In that operation, the actual value of the pump frequency and the recoil term both cancel out, and the shift is

$$\nu_0 (B) - \nu_0 (0) = \nu_{jk} (B) - \nu_{jk} (0) - \lambda/\lambda [\nu_{ij} (B) - \nu_{ij} (0)]. \quad (35)$$

The frequency differences in the right-hand side of Eq. 35 are conveniently expressed writing the linear $B$-dependency of the energies as a function of the $\hat{m}$ quantum numbers in the three involved sublevels, characterised by the three indices $i$, $j$, and $k$, so that the linear Zeeman shift factor is

$$\nu_0 (B) - \nu_0 (0) = \left[ \frac{\lambda}{\lambda} \hat{g}_1 (i) \hat{m}_1 (i) - \frac{\lambda}{\lambda} \hat{g}_2 (j) \hat{m}_2 (j) + \hat{g}_3 (k) \hat{m}_3 (k) \right] \mu_B \quad (36)$$

where $\hat{g}_1$, $\hat{g}_2$, and $\hat{g}_3$ are the $g$-factors ($g_P$ for $^3$He, $g_S$ or $g_J$ for $^4$He) of the involved sublevels of the $2^3S$, $2^3P$, and $3^3S$ states, respectively, and $\hat{m}_1$, $\hat{m}_2$, and $\hat{m}_3$ their angular momentum projections. Note that the signs in the weights affecting the contributions of the energy shifts in the three states in Eq. 36 are those obtained for the co-propagating beam configuration; different weights would be derived for a counter-propagating scheme, not used in this work.

Equation 36 can be explicitly written for any pump and probe line component, using the values of $g$-factors listed in Table 1 and immediately after Eq. 4. For the simple scheme involving the $2^3P_0$ state of $^4$He depicted in Fig. 3a it reduces to

$$\nu_0 (B) - \nu_0 (0) = \left[ \frac{\lambda}{\lambda} m_S + \star m_S \right] g_S \mu_B. \quad (37)$$

For a $\pi$-polarised pump ($m_S = 0$ is selected), a triplet of absorption lines can potentially be observed (depending on the probe polarisation) with the usual $\star g_S \mu_B$
splitting factor (28 MHz/mT). Conversely, for a fixed π-polarisation of the probe (\( n_S = 0 \)), three velocity classes can potentially be excited (depending on the pump polarisation) and a triplet of absorption lines can also be observed, with an enhanced splitting factor, \( g_{SB} \lambda / \lambda (42.9 \text{ MHz/mT}) \). Altogether, when all possible pump and probe polarisations are considered, nine lines may be observed with unevenly spaced shift factors spanning ±70.9 MHz/mT.

### 3 Experimental setup and protocol

The main elements of the double optical resonance experiments lying in the same plane are displayed in Fig. 4: the pump and probe lasers, the co-propagating beams overlapping in the experimental cell \( C_{\text{exp}} \), the light polarisation and intensity control elements, and the saturated absorption setup which uses part of the pump beam and the ancillary cell \( C_{\text{lock}} \). They are successively described in this section, along with off-plane light beams and elements. The setup had a 1.8 \times 0.5 \text{ m}^2 footprint.

**Cells**—Experiments were performed at room temperature with helium gas in sealed cylindrical Pyrex glass cells. Their dimensions ranged from 3 to 12 cm in length and from 1.5 to 5 cm in diameter. The filling pressures ranged from 0.13 to 67 mbar of \(^3\text{He} \), \(^4\text{He} \), or isotopic mixtures. The cells had been made in-house for previous experiments according to a standard cleaning and filling protocol [17, 24]. They were positioned with a small tilt angle with respect to the light beams to avoid large overlap from the reflections on the uncoated cell windows. A weak RF discharge was used to populate the \( 2^3\text{S} \) state with external wire electrodes arranged according to the cell shape. The RF frequency was of order 2–3 MHz or 20–25 MHz, depending on cell size and gas pressure. Low RF frequencies usually provided stable operation for weak discharges and lower RF interference noise whereas higher frequencies were used for easier discharge ignition at the lowest gas pressures. \( C_{\text{exp}} \) was located at the centre of the coil sets and \( C_{\text{lock}} \) was 7.5 cm lower and further along \( z \) so as to practically eliminate RF crosstalk and couplings of discharge intensities between cells.

**Magnetic-field coils**—A set of six square coils, similar to that used in Ref. [30], was used to apply an axial magnetic field component \( B_z \) up to 4 mT, with a computed relative field standard deviation (SD) of \( 7 \times 10^{-5} \) over the probed volume in a 12-cm-long experimental cell. Two pairs of rectangular coils were used to apply field components transverse to the light beams, \( B_x \) (vertical) and \( B_y \) (horizontal, perpendicular to the plane of Fig. 4) with magnitudes up to 2 mT and computed relative SD of \( 1.3 \times 10^{-3} \) and \( 2 \times 10^{-3} \), respectively, over the same probed volume. Details on the coil design, construction, and computed field maps can be found in Ref. [31]. The coil sets were used to cancel the unwanted Earth’s field components and to control the total field direction and magnitude in the experiments.

**Pump laser**—The laser source for the 1083 nm pump beam was a distributed Bragg reflector (DBR) diode laser (DL) with on-chip thermistor and thermo-electric cooler (Toptica LD-1082-0070-DBR) delivering 50 mW at our maximum operating current. The beam was collimated using an aspheric lens of 8 mm focal length. It was linearly polarised, with a polarisation vector along \( x \) (perpendicular to the plane of Fig. 4). The pump beam was mapped at the cell location (0.2 \times 0.7 \text{ cm}^2 \text{ FWHM}) using a beam profiler (Thorlabs BC106N). Its on-axis maximum intensity inside the cell (\( \approx 90 \text{ mW/cm}^2 \)) was fairly uniform over the area selected by the cropping diaphragm (labelled D in Fig. 4). It was evaluated taking into account the reflectance of the cell windows (typ. 9% per window). The sample transmittance for various pump intensities was inferred from pairs of pump power measurements after the cropping diaphragm with RF discharge on and off.

In some experiments, the pump laser was used in free-running operation, with its frequency set by the choice of the operating current and temperature (regulated using an in-house DL controller). To conveniently monitor pump absorption by the He gas in the cells, the discharge RF amplitudes could be modulated (around 80 Hz). Corresponding lock-in demodulation of signals from \( P_1 \) and \( P_2 \) was then performed and outputs similar to the sketch in Fig. 3b (see Fig. 11) were obtained for \( P_1 \) during pump frequency scans.

In other experiments, the pump laser frequency was locked on a helium transition using a standard satu-
rated absorption scheme applied to gas contained in the ancillary cell C_{lock} (Fig. 4). A small modulation of the DL current at 1.5 kHz was used to induce a pump frequency modulation and lock-in demodulation provided a suitable error signal for a servo loop. The use of a quarter-wave plate allowed for convenient separation of the reflected beam by the PC-QW element after a double pass in C_{lock}. Moreover circularly polarised light yielded fewer (or single) line components in Zeeman spectra and therefore better frequency locking schemes. For most experiments with a locked pump laser, a mixture cell (with 1.07 mbar \(^3\)He and 0.53 mbar \(^4\)He) was used to lock on any of the three \(^4\)He lines or on the \(^3\)He C\(_8\) line.

**Probe laser**—The laser source for the 706.5 nm probe beam was an extended cavity diode laser (ECDL, Toptica DL 100 L). We used a second in-house DL controller to manage the DL temperature and current, and a computer to control this current and the high voltage applied to the piezo actuator of the grating mount. Empirically determined joint linear variations of these current and voltage allowed for mode-hop-free tuning across the full helium spectra (\(> 40 \text{ GHz}, \text{see Fig. 2c}\)). This range exceeds the nominal specifications (21 GHz), which was sometimes convenient for wide frequency scans. However, large amplitude noise was often observed near the boundaries of the scanned spectra and such large frequency intervals involved large changes in the DL current, thus in the probe amplitude. Therefore, suitable DL temperature adjustments were routinely used to centre the scanned intervals on regions of interest in the absorption spectra. For a DL current close to the maximum operating value at the lower frequency, the probe power decreased by about 30% at the higher frequency of typical 15 GHz scans.

**Probe wavelength measurement**—The probe wavelength was monitored using a Fizeau wavemeter (HighFinesse WS6-600). Its fibre input (not shown in Fig. 4) collected light deflected by the beam splitter BS\(_2\) in a horizontal plane. The recorded vacuum wavelength values \(\lambda^*\) were used to compute frequency offsets \(\nu\) using our nominal offset (\(\nu^*(4)/h\) in Table 2) and the published vacuum wavelength values \(\lambda_0\) of the \(*D_0\) transition:

\[
\nu = \nu^*(4)/h + c(1/\lambda - 1/\lambda_0),
\]

\[
\lambda_0 = 706.76568 \text{ nm}.
\]

The wavemeter absolute accuracy was 600 MHz, but its specified resolution (20 MHz) and its readout resolution (10^{-5}\text{ nm}, hence 6 MHz at the probe wavelength) could yield fairly precise measurements. Characterisations of its measurement errors were performed and are described in Appendix A. As expected for a Fizeau interferometer, the readout wavelengths remained quite stable, with corresponding frequency drifts not exceeding 10 MHz during a relevant scan time interval (10 to 30 s) when the probe laser frequency was not swept.

In spite of this fair time stability of the readout data, recordings performed for swept lasers frequencies indicated that instrumental errors of tens to hundreds of MHz varied with the laser frequency, an observation similar to that recently reported for a high-accuracy device [33]. Such frequency dependent errors induced local scaling errors, not exceeding \(\pm 5\%\), affecting for instance the obtained line widths or areas. These errors were usually reproducible for successive repeated recordings, but lost their correlations for recordings made hours or days apart. Averaging results of distant measurements may therefore be used to reduce the effect of the wavelength-dependent readout errors on line width and line splitting measurements.

**Probe light detection**—A general-purpose optical polarimeter (not shown in Fig. 4) included a polarising beam splitting cube (and an additional polariser in the deflected channel) for separation of the horizontal and vertical polarisation components of the probe light (H and V, aligned with x and y in the measurement cell, respectively) [34,35]. The split beams were coupled by lenses of 30 mm focal lengths into 1-mm core multimode plastic fibres that channelled light to Si PIN photodiodes (Siemens BPW34), and photocurrents were suitably converted to voltages. This light collecting scheme for the photodetectors P\(_H\) and P\(_V\) provided a high directional selectivity and made detection immune to stray light. It was used for the 1083-nm-sensitive detector P\(_1\) (Siemens BP104F) as well, whereas a large area photodiode (Centronic OSD50-4X) was directly placed on the path of the pump for detector P\(_2\).

The direction of the linear polarisation of the probe light was rotated using a half-wave plate HW. Only horizontal or vertical polarisations in cell C_{exp} (characterised by null signals from detectors P\(_V\) and P\(_H\), respectively) were used in standard experiments. They were fully preserved by the dichroic mirrors DM\(_1\) and DM\(_2\) and were thus well defined in the experimental cell. Therefore, for a transverse applied magnetic field along the direction of the linear light polarisation (\(\pi\)-polarised light), only \(\Delta m = 0\) transitions were excited or probed. Otherwise \(\sigma\)-polarised light was obtained for field directions perpendicular to that of the light polarisation (with equal intensities of \(\sigma_+\) and \(\sigma_-\) components), driving \(\Delta m = \pm 1\) transitions. Combinations of \(\pi\) and \(\sigma\) polarisations, achieved for slanted orientations of the field in the transverse plane, were used in some experiments.

**Data acquisition**—For a standard experiment, the pump frequency was fixed and locked to an atomic transition unless otherwise specified. The pump intensity was varied using attenuators. The pump intensity in the sample and that absorbed by the sample
were inferred from power measurements with discharge off and on (performed at low pump intensities using the photodetector P<sub>2</sub>). For fixed discharge and pump settings, the probe laser frequency was linearly swept over a chosen interval, either in single pass or periodically for several scans. Probe frequency sweep rates of 200 MHz/s or slower were normally chosen, depending on the linewidths of the probed transitions.

During recordings the pump beam intensity was modulated (at 1.5 kHz) using a rotating wheel optical chopper. The signals from the photodetectors P<sub>H</sub> and P<sub>V</sub> were demodulated using digital lock-in amplifiers (Signal Recovery / EG&G 7220). An output time constant of 20 ms (6dB/octave) was usually chosen. The raw photodetector signals were also integrated with simple RC networks to filter out the modulation, and both the integrated and the demodulated signals were digitally recorded. A 16-bit USB data acquisition ADC module (Data Translation DT9816) with a 1-kHz sampling rate was managed by an in-house program that simultaneously controlled the probe laser frequency via a 12-bit DAC module (DT9813). Decimation of the over-sampled data was performed in post-processing. Digital or aliased noise resulting from the digital data recording were thus made negligible. The variations of the probe wavelength (vacuum wavelengths versus time) were simultaneously recorded using the proprietary software of the wavemeter. The probe beam was transiently blocked at the beginning of each recording, which provided a time stamp used in data processing to accurately align optical and wavelength data and eliminate time.

### 4 Results

#### 4.1 Examples of double optical resonance spectra

Illustrative examples of double optical resonance experimental spectra are compared in this section. The probe transmittance \( \mathcal{T} \), which can be directly linked with the atomic density distributions of velocities in the 2<sup>3</sup>P state sublevels (Sects. 2.2 and 2.3), was computed for each photodetector (P<sub>H</sub> or P<sub>V</sub>) from the ratio of demodulated signal magnitudes, \( V_{\text{RMS}} \), and averaged signal levels, \( V_{\text{av}} \), as

\[
\mathcal{T} = \frac{1 - \beta V_{\text{RMS}}/V_{\text{av}}}{1 + \beta V_{\text{RMS}}/V_{\text{av}}}.
\]  

(40)

This approach is reminiscent of that used in the case of weakly modulated densities of absorbers [24, App. A], but in this experiment the density of 2<sup>3</sup>P-state absorbers was fully modulated with a near-square time pattern. For negligible rise and fall times of the pump intensity the coefficient in Eq. 40 would be \( \beta = \pi/(2\sqrt{2}) \). The effect of the finite transition times introduced by the pump beam chopper was assessed on the signals from the pump photodetector P<sub>2</sub>. As a result the corrected value \( \beta = 1.03\pi/(2\sqrt{2}) \) was used.

In order to conveniently compare recorded transmittance spectra to theoretical expectations, frequency offsets (i.e. shifts from the \( ^5\text{P}_1 \) line) were inferred from wavelength measurements in two steps. Recorded wavelength data were first suitably processed to reduce spurious noise and interpolation of the processed data was used to ascribe a wavelength value to each computed transmittance value. The corresponding experimental frequency offset was then obtained using Eq. 38.

Figure 5 displays examples of double optical resonance data selected to show the effect of the gas composition and pressure on the observed absorption spectra. For the top row (Fig. 5a–c), the pump selectively excited each of the three \( J \)-levels of the 2<sup>3</sup>P state of \(^4\text{He} \). Absorption of the probe predominantly occurred from the pumped level, as expected. The weak absorption signals corresponding to atoms in the unpumped levels (note the 10-fold gain factor on one side of the broken axis in Fig. 5a–c) result from small population transfers induced by \( J \)-changing collisions during the 2<sup>3</sup>P state radiative lifetime. For the experiments of Fig. 5d, e, the 2<sup>3</sup>P<sub>0</sub> levels of \(^4\text{He} \) and \(^4\text{He} \) were excited in the same cell using the well-resolved D<sub>0</sub> and C<sub>8</sub> pumping transitions, respectively. Narrow lines still correspond to absorption by atoms and isotopes selectively promoted to the pumped levels, and signals corresponding to absorption by atoms in the un-pumped levels are more salient than at lower pressure, as expected. With the large signal-to-noise ratio available in these measurements (the typical standard deviation in the baselines was 5 to \( 9 \times 10^{-5} \)), the transition frequencies could be inferred with a small statistical uncertainty (\( 0.1 \) – 1 MHz). The agreement with the computed frequency offsets (depicted as vertical bars below each spectrum) was usually fair but significant discrepancies (tens to hundreds of MHz) were often found (for instance in Fig. 5a for the main line and in Fig. 5b, c for weak lines). This issue is discussed in Appendix A and illustrated in Sect. 4.4.

The high-pressure absorption data in Fig. 5f show no evidence of narrow features, but resemble a computed Doppler-broadened spectrum (Fig. 2c). However, attempts (not shown) to fit absorption data using computed spectra revealed sizeable deviations in line shapes unless Voigt (rather than Gaussian) profiles were used, as expected, and a slightly weaker weight was ascribed to the 2<sup>3</sup>P<sub>0</sub> population.

The total populations of the various levels of the 2<sup>3</sup>P state and population-transfer collision rates may be inferred from areas of line components in such data. Globally, 80 to 90% of the 2<sup>3</sup>P population lied in the pumped state and velocity class for the low-pressure experiments (Fig. 5a–c). In contrast, only 25% remained in the pumped 2<sup>3</sup>P<sub>0</sub> velocity class for the 1.6 mbar gas mixture (Figs. 5d, e), and 10% (close to the thermalised limit of 1/9) of the atoms were detected in the un-pumped 2<sup>3</sup>P<sub>0</sub> level for a 67 mbar \(^3\text{He} \) gas submitted to excitation to the 2<sup>3</sup>P<sub>2</sub> and 2<sup>3</sup>P<sub>x</sub> levels (Fig. 5f).

The \( B = 0.16 \) mT applied field was transverse and such that \( B_y = 2B_x \) for the experiments on low-
the different transmittance and frequency scales for the top high-pressure from Fig. 2a, b are displayed below each spectrum. Note pump intensity incident on the cell was 55 mW/cm$^2$ or 0.1 W/cm$^2$.

probe detuning from C is indicated in each box. Transmittance data are plotted vs. to: DOP-induced build-up of atomic alignment in the 2$^3$S state ($σ_+$, $σ_-$, and $π$) which efficiently prevented OP-induced build-up of atomic alignment in the 2$^3$S state. This yielded an increase in both pump and probe absorption signals by a factor $∼2$ compared to field orientations corresponding to a $π$- or $σ$-polarised pump having the same total intensity. For experiments in cells containing sizeable amounts of $^3$He (e.g., Fig. 5d-f), ME collisions with spin-1/2 ground state atoms efficiently prevented alignment to occur in the 2$^3$S state and absorption of pump and probe light was indeed observed to be almost unaffected by the direction of the applied field.

4.2 Examples of Zeeman-split spectra

Figure 6 displays probe absorption spectra obtained in an applied longitudinal field when the 2$^3$P$_0$ state of $^3$He (Fig. 6a) or $^4$He (Fig. 6b) was pumped. The Zeeman shifts derived in Sect. 2.4 were used to compute the expected line positions represented below the spectra.

The $^3$He line component shifts were obtained using Eq. 35. For $^3$He pump and $^8$C$_9$ probe all $F$-values are equal to 1/2 and only two excitation paths $m_1 \rightarrow m_2 \rightarrow m_3$ are allowed for $σ$-polarised light: $±1/2 \rightarrow ±1/2 \rightarrow ±1/2$, yielding a doublet with shift factors computed using Eq. 36 and noted $±β_9 = ±44.18$ MHz/mT. For the $^8$C$_9$ probe, $F = 3/2$ in the upper level and four excitation paths are allowed, yielding a barely split strong doublet ($m_3 = ±3/2$, with shift factors $±β_9,β_4 = ±2.54$ MHz/mT, $T_{17,4} = 0.0937$) and a weak doublet ($m_3 = ±1/2, ±β_0,1, = ±34.84$ MHz/mT, $T_{17,2} = T_{17,4}/3$). The recorded spectra exhibit the corresponding features in Fig. 6a, and sums of Lorentzian profiles (the red lines) accurately reproduce the experimental data. Four parameters were used in the fitting function $F_S$ for $^8$C$_9$ spectra, yielding values for the total line area $A_{S\text{fit}}$, the common width $W_{S\text{fit}}$, the field strength $B_{S\text{fit}}$, and a frequency offset $δ_{S\text{fit}}$.

$$F_S (ν_0) = A_{S\text{fit}} [L_{W\text{fit}} (ν_0 - ν_8 + δ_{S\text{fit}} + β_8 B_{S\text{fit}})] + L_{W\text{fit}} (ν_0 - ν_8 + δ_{S\text{fit}} - β_8 B_{S\text{fit}})]/2.$$  

The fit value $B_{S\text{fit}}$ was found in good agreement with $B_z$, the 0.5% difference being lower than the possible scaling errors in frequency scale discussed in Appendix A. Three parameters were used in the fitting function $F_S$ for $^8$C$_9$ spectra: $A_{S\text{fit}}$, $W_{S\text{fit}}$, and $δ_{S\text{fit}}$. 

[Fig. 5 Experimental double optical resonance spectra in low-pressure $^4$He (a–c), an isotopic mixture (d, e), and high-pressure $^4$He (f). The gas pressure and composition is indicated in each box. Transmittance data are plotted vs. probe detuning from $^3$C. Note the 4.5-GHz-long break in the frequency axis of panel a.]

[Fig. 6 Double optical resonance spectra displaying combined Zeeman line splittings ($σ$-polarised pump and probe beams). a Pump tuned to $^8$C$_9$ (55 mW/cm$^2$), $B_z = 3.14$ mT, 0.27+0.27 mbar isotopic mixture. b Pump tuned to $^4$D$_0$ (100 mW/cm$^2$), $B_z = 3.68$ mT, 0.133 mbar $^4$He gas. Experimental data (black), Lorentzian fits (red, Eqs. 41–43), and residues (teal) are displayed. The bars below the spectra are the computed positions and transition matrix elements of the line components. Note the 4.5-GHz-long break in the frequency axis of panel a.]

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while the nominal applied field $B_z$ was used since the poorly resolved lines could not reliably yield both field and width fit values.

\[
F_0 (\nu_0) = A_{0\text{fit}} [3L_{W0\text{fit}} (\nu_0 - \nu_9 + \delta_{0\text{fit}} + \beta_{03} B_z) \\
+ 3L_{W9\text{fit}} (\nu_0 - \nu_9 + \delta_{9\text{fit}} - \beta_{93} B_z) \\
+ L_{W0\text{fit}} (\nu_0 - \nu_9 + \delta_{0\text{fit}} + \beta_{01} B_z) \\
+ L_{W9\text{fit}} (\nu_0 - \nu_9 + \delta_{9\text{fit}} - \beta_{91} B_z)] / 8.
\]

(42)

The frequency offsets $\delta_{0\text{fit}}=17$ MHz and $\delta_{9\text{fit}}=124$ MHz both lied within the precision of the frequency measurements (600 MHz). They differed from a common expected offset associated with the use of circularly polarised light to set or lock the pump laser frequency (Fig. 4). Such difference is discussed in more detail in Sect. 4.4. The linewidths $W_{0\text{fit}}=119$ MHz and $W_{9\text{fit}}=115$ MHz, each with a 2 MHz statistical uncertainty and a few % of uncertainty arising from possible scaling errors, are in satisfactory agreement. Finally, the ratio $A_{0\text{fit}}/A_{9\text{fit}}=1.262$ of the line areas parameters was found to be consistent with the expected value, $(T_{17.4} + T_{17.2}) / T_{17.5} = 1.246$.

The $^4\text{He}$ shifts in Fig. 6b were obtained using Eq. 36. Four double resonance excitation paths through $m_J=0$ are allowed for $\sigma$-polarised light: $m_S=\pm 1$ to $m_S=\pm 1$, yielding one doublet with shift factors $\pm \beta_{03}=\pm 70.91$ MHz/mT, and $m_S=\pm 1$ to $m_S=\pm 1$, yielding another doublet with shift factors $\pm \beta_{03}=\pm 14.91$ MHz/mT. A fitting function $F_0$ for $^4\text{D}_0$ with parameters $A_{0\text{fit}}$, $W_{0\text{fit}}$, $B_{0\text{fit}}$, and $\delta_{0\text{fit}}$ was used to model the Zeeman-split line in Fig. 6b:

\[
F_0 (\nu_0) = A_{0\text{fit}} [L_{W0\text{fit}} (\nu_0 - \nu_0^{(4)} + \delta_{0\text{fit}} + \beta_{03} B_{0\text{fit}}) \\
+ L_{W0\text{fit}} (\nu_0 - \nu_0^{(4)} + \delta_{0\text{fit}} - \beta_{03} B_{0\text{fit}}) \\
+ L_{W0\text{fit}} (\nu_0 - \nu_0^{(4)} + \delta_{0\text{fit}} + \beta_{03} B_{0\text{fit}}) \\
+ L_{W0\text{fit}} (\nu_0 - \nu_0^{(4)} + \delta_{0\text{fit}} - \beta_{03} B_{0\text{fit}})] / 4.
\]

(43)

Again, the fit value $B_{0\text{fit}}$ (1.5% lower than $B_z$) and the frequency offset $\delta_{0\text{fit}}=-138$ MHz were in line with expectations. The linewidth $W_{0\text{fit}}=90$ MHz, which was obtained with a 1 MHz statistical uncertainty, cannot be directly compared at this stage with that obtained at a different pressure, for the other isotope, and with a different pump intensity. We still notice that, for the two $^2\text{S}$-$^2\text{P}_0$ pumping schemes considered here, two narrow velocity classes with identical widths can be observed for the atoms detected in the $^2\text{P}_0$ level (one class for each of the two magnetic sublevels in $^3\text{He}$, two classes with opposite mean velocities in $^4\text{He}$ when the pump is tuned to the resonant atomic frequency for $B_z=0$). Therefore, a single width parameter can be used in all fit functions, which yields robust fit results even if the Zeeman components are ill-resolved. We have thus a suitable tool for fits of narrow (Doppler-free) lines, from which areas and widths can be reliably extracted. The situation would be more complex for pumping schemes addressing different sublevels with different pumping rates, for instance.

4.3 Effect of pump intensity

The influence of pump light intensity on probe absorption signals was studied for several conditions of gas composition and pressure, as well as of pump light polarisation and frequency. The strength of probe absorption was characterised by the areas of the narrow spectral lines corresponding to directly pumped atomic sublevels in each experiment. The areas were usually obtained from fit parameters of suitable model functions. They were checked to be consistent with those obtained from numerical integration whenever spectral shapes and signal-to-noise ratio allowed for it, for instance at high pump intensity and low gas pressure.

Figure 7a displays data obtained in different cells for $\pi$-polarised pump and probe light, plotted as functions of average pump intensities $\langle I_p \rangle$ (which were computed assuming exponential decay along the pump path). A wide range of area values was obtained at fixed pump intensity, as could be noticed for instance in Fig. 5. The solid lines in Fig. 7a are the low-intensity areas of Eq. 29 computed for the experiments made in the low-pressure cell using the low-intensity transmittance $T_0$. They agree with the corresponding data (solid triangles) within 10% below a few nW/cm², and the differences at higher intensities emphasize the strong loss of efficiency in populating the probed $^2\text{P}_3$ state over most of the pump intensity range. All area values tend to saturate at large pump intensity, which correlates with the decrease of pump absorption plotted in Fig. 7b for the corresponding experiments. It is worth noticing that the decrease with $I_p$ was faster for pure $^4\text{He}$ (all solid symbols), in which atomic alignment was produced in the $^2\text{S}_1$ state by $\pi$-polarised pump light. Optical pumping reduced absorption in that case, but it was efficiently hindered by ME collisions when $^3\text{He}$ or isotopic gas mixtures were used. Of course, enforcing equal intensities of the three pump light polarisation components (see Sect. 4.1) was another way to avoid optical pumping effects and to enhance absorption at large $I_p$ in pure $^4\text{He}$.

Figure 7c combines the results obtained for measured areas (data of Fig. 7a) and measured pump intensity differences (between input and output of the gas samples), $\Delta I_p$, as suggested by Eq. 26. The experimental errors for the areas mainly arose from frequency scaling errors (up to $\pm 5\%$) discussed in Appendix A. For the weakest absorptions at high $I_p$ the accuracy on $\Delta I_p$ was rather poor as a result of large interference from the RF discharge (see a corresponding worst-case error bar in Fig. 7c). Allowing for these errors, the computed ratios were found to be quite independent of pump intensity for each sample, as expected. Moreover, their values were consistent with the expectation of Eq. 26, with fractions $R$ of atoms close to 1 at 0.13 mbar and
4.4 Feasibility of Doppler-free spectroscopy

Following the conclusion of Appendix A, series of repeated scans of the $^2\!^3P_0$–$^3\!^3S$ transitions in isotopic gas mixtures were performed with the pump frequency locked to the $C_8$ or the $D_0$ line. The experiments were performed in null field to avoid Zeeman line shifts. Indeed, with linear pump and probe polarisations, Zeeman splittings (such as observed in Fig. 6) were not expected to affect centre-of-line positions. In contrast, the saturated absorption beam was circularly polarised and therefore the pump frequency would have been Zeeman-shifted proportionally to any $B_z$ field. To avoid this, the three Earth’s field components were compensated and in particular $|B_z| < 2 \mu T$, resulting in a worst-case frequency shift below 0.1 MHz in these experiments (Eq. 36).

Figure 8a displays the time variations of the probe wavelength and absorption spectra for six back and forth scans, for a $C_8$-tuned pump. For each scan in...
Table 7 Statistical data for the results compiled in Fig. 8. Columns 2 to 4 list the differences between the averaged fit frequencies and the computed values, the standard deviations and the standard. Ns is the number of scans used for each average. The last line contains the data for the $^3\text{He}$ hyperfine splitting measurements (see text and Fig. 8e).

| Line name | Av.-th. (MHz) | SD (MHz) | SE (MHz) | Ns |
|-----------|---------------|----------|----------|----|
| $^3\text{C}_8$ | $-38.1$ | $177$ | $18.5$ | $92$ |
| $^3\text{C}_9$ | $-39.7$ | $144$ | $15$ | $92$ |
| $^3\text{D}_0$ | $2.0$ | $63.6$ | $7.5$ | $72$ |
| $^3\text{C}_9 - ^3\text{C}_8$ | $1.6$ | $76.5$ | $8$ | $92$ |

a recording the $^3\text{C}_8$ and $^3\text{C}_9$ absorption lines were fit using Lorentzian profiles, yielding sets of fit frequencies, linewidths, and statistical uncertainties.

Figure 8b displays the fit frequencies of the $^3\text{C}_8$ transition in Fig. 8a scans together with those from three more recordings made on the same day. The frequency drifts between scans of a recording or between recordings were on the same order of magnitude as the typical linewidth ($\Delta \nu = 120$ MHz, vertical bar in Fig. 8b). The statistical uncertainty on fit frequencies ($\sim 1$ MHz) was smaller than symbol sizes.

The results of all measurements for the $^3\text{C}_8$ transition are compiled in Fig. 8c, in which the symbol positions and error bars stand for the averages and standard deviations of fit values obtained in each recording (the four recordings used in Fig. 8b yield the four data points of day 2). The results in Fig. 8c–e correspond to 29 recordings made on four different days using two cells with two gas mixtures: $0.27+0.27$ mbar $^3\text{He}+^4\text{He}$ on consecutive days 1 and 2, and $0.13+0.4$ mbar on days 3 and 4 one week later. The pump was tuned to $^3\text{C}_8$ to excite $^3\text{He}$ atoms (Fig. 8c–e, 17 recordings) or to $^3\text{D}_0$ to excite $^3\text{He}$ atoms (Fig. 8f, 12 recordings).

With similar plotting conventions Fig. 8e displays the differences in fit frequency for the $^3\text{C}_8$ and $^3\text{C}_9$ lines computed for each scan recorded with $^3\text{C}_8$ pumping. The in-recording standard deviations (the error bars in Fig. 8e) and the scatter of average values of the line splittings are somewhat reduced compared to the absolute frequency measurements, but the frequency-dependent wavemeter errors were not eliminated in the process.

Table 7 lists the quantitative results of this series of measurements in terms of deviations from the computed values. The inferred hyperfine splitting in the $3^3\text{S}$ state of $^3\text{He}$ and isotope shift of the 706.5 nm transition are in good agreement with expectations in spite of the large and frequency-dependent readout errors of the wavemeter used in these experiments that make this instrument ill-suited for accurate spectroscopic measurements. The standard errors obtained for repeated measurements suggest that the measurement errors were in fact efficiently averaged.

5 Discussion

Calculations of the level structures and optical transition details for the 1083 nm transition of Ref. [17] were extended to the 706.5 nm transition. Explicit expressions of Zeeman shifts of the $2^3\text{S}$, $2^3\text{P}$, and $3^3\text{S}$ levels involved in double optical resonance are given in this article for the low-field limit, but this extension is indeed valid for arbitrary magnetic field using straightforward changes in the numerical computations of Ref. [17]. Regarding double optical resonance schemes, quantitative links were derived between integrated probe absorption spectra at 706.5 nm and atomic densities in the $2^3\text{P}$ state or pump absorption.

As regards line shapes, the low-pump-intensity limit was derived and the power broadening effects on velocity distributions in the $2^3\text{S}$ and $2^3\text{P}$ states were briefly discussed in Sect. 2.3. They would indeed automatically induce a related broadening of probe absorption lines. Additional broadening of the probe lines resulting from an intense pump may be expected for this 3-level ladder-type excitation scheme. Various theoretical approaches have been used in similar situations (e.g., [36–38]). A pump-induced increase of the damping rate of the probe transition may be expected, with changes in the line shapes expected from Rabi oscillations driven by an intense pump in a dressed-atom approach. Such effects are not believed to play a significant role in our experiments where strong optical saturation was not achieved, but a dedicated study at higher pump light intensity would probably benefit from the use of optical probes on both optical transitions.

A simple experimental scheme was used with collinear pump and probe beams combined using dichroic mirrors. This restricted the beam light polarisations to being linear and either s- or p-polarised (horizontal or vertical polarisations in our setup). This was required to avoid strong modifications of the beam polarisations, which are well known for reflections on dichroic mirrors [39] and were observed in this work for the transmitted beam as well. Experiments requiring arbitrary beam polarisations could advantageously be performed with pump and probe beams propagating with a small angle to avoid the need for compensation of the effects of dichroic mirrors, at the expense of a loss of compactness of the system and of imperfectly cancelled Doppler broadening.

Double optical resonance spectra recorded with our simple scheme over a wide range of pressures displayed very clear features, with narrow Doppler-free lines below a few millibars. In that case complex Zeeman splittings resulting from the interplay of the Zeeman shifts and pump-induced velocity selection were observed and found to agree well with expectations. This provides a basis for reliable analyses and fits of the observed spectra in applied magnetic fields, in which line shapes, widths, and positions of individual components can be reliably determined even in poorly resolved Zeeman structures. Conversely, above a few tens of millibars, broad thermal-like spectra provided
evidence of efficient velocity-changing collisions during the radiative lifetime of the 23P state. The velocity-selective character of the pump was essentially lost but the tremendous increase in signal-to-noise ratio of this approach, compared to conventional absorption or fluorescence techniques, still makes our pump-probe scheme very useful. And indeed, for the studies in the intermediate pressure regime which have motivated these experiments, analyses of recorded spectra for suitable combinations of pump and probe frequencies and polarisations can be expected to provide a wealth of useful data. The reported observations of the influence of pump intensity on signal amplitudes already highlight the pressure dependence of the fractional transfer of populations, both in pure 4He and isotopic mixtures. It also provides values for suitable pump intensities in various situations, with large enough probe absorption and moderate saturation broadening.

Incidentally, an intrinsic frequency-dependent measurement error of a Fizeau-type wavemeter was evidenced and characterized. The resulting limitation on the precision of spectroscopic measurements on the Doppler-free 706.5 nm transition was evaluated, with a 10–20 MHz statistical error on line positions in our feasibility study. This already yields improved determinations of the hyperfine structure constant and isotope shift of this transition compared to prior results [12,13,16]. However, obtaining spectroscopically useful results with accuracies not exceeding sub-MHz statistical errors in a single scan would require a more accurate wavelength measurement instrument or method in order to settle, for instance, the 0.55 MHz discrepancy between the published isotope shift differences of Eqs. 6 and 7. Suitable measurements could be performed using, for instance, a more accurate model by the same manufacturer (with a 60-fold smaller stated error) or a stable interferometer (of the kind used in Ref. [16] for He absorption spectroscopy at 706.5 nm or in Ref. [40] for the determination of the He isotope shift at 1083 nm).

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Author contributions

G.T and P.J.N. designed and initiated the project. A.D. performed most of the reported experimental work and data analysis. P.J.N. performed the calculations and wrote the manuscript with input from all authors.

Data Availability Statement This manuscript has no associated data or the data will not be deposited. [Authors’ comment: The datasets generated and analysed during the reported study are available from the corresponding author upon reasonable request.]

Appendix

A Characterisations of wavemeter measurement errors

Wavemeters based on Fizeau interferometers have a resolution and stability significantly superior to their absolute accuracy. This feature has been used by several groups to precisely lock lasers at fixed frequencies using devices that have the highest available accuracy (10 MHz instead of 600 MHz in our experiments) [41,42]. In this Appendix we report on checks of the readout stability of our wavemeter at fixed laser frequency, then examine an ill-documented feature of readout errors for frequency-swept lasers, and illustrate the influence of such errors on measurements of well-known line component splittings for the 1083-nm transition.

A.1 Wavemeter readout stability

We have evaluated the time stability of measurement errors by monitoring readout drifts at fixed laser frequencies, for the un-swept probe laser sitting on a narrow double optical resonance line, or for the locked pump laser (in that case, the photodetector P2 was replaced with the collimator of the wavemeter input fibre). Slow readout drifts were observed in both cases, and rapid changes were observed when the wavemeter input fibre was bent or moved or when the light injection into the fibre was geometrically modified. Figure 9 displays an example of long-term recording of the wavemeter readout λ with the pump locked on the D2

![Fig. 9 Recording of wavemeter data for the 1083-nm pump DL locked on the 4He D2 line (of wavelength λ2; see text). The inset is a ten-fold blow-up of part of the recording, with raw data (black, 10 ms sampling) and smoothed data (red, 20-point running averages) achieving a sub-MHz precision]
The difference of wavelengths of the damped oscillations following each turning point was of order 1.6 to 2.6 pm, i.e. 10 to 70 MHz in this example, which was consistent with the instrument accuracy (600 MHz). Short-term drifts (see the blow-up in Fig. 9) during typical scan times in double optical resonance experiments (10 to 30 s) did not exceed 10 to 20 MHz, with a sub-MHz precision for smoothed data. The sudden noise increase and readout jump near the end of the recording were induced by small movements of the wavemeter input fibre.

A.2 Wavelength measurements for repeated scans of pump or probe lasers

Most recordings exhibit smooth variations (Fig. 10a) but apparent jumps were sometimes observed (Fig. 10b). The damped oscillations following each turning point in the triangular sweeps were actual DL temperature (and thus frequency) oscillations but the other features were variations in the wavemeter error similar to those recently reported for a high-accuracy device [33]. The use of wavemeter data instead of the true wavelengths when processing absorption data therefore induced local scaling errors, given by the reduced derivatives of the deviations from linear variations, plotted as the upper traces in Fig. 10a,b. These scaling errors typically did not exceed ±5%.

When the probe laser wavelength was recorded a clear hysteretic response was observed (Fig. 10c). Subtracting a linear function of the control parameter (Fig. 10d) did not suggest a reliable parametrisation for this behaviour, which was attributed to the mechanical response of the piezo actuator. We could therefore not avoid the local scaling errors resulting from the use of wavemeter data, and simply assumed that they had similar magnitudes at pump and probe wavelengths. These frequency-dependent errors were usually stable for successive recordings, which is evidenced by the rather similar traces within Fig. 10a or b. In contrast they lost their correlation for recordings made hours or days apart, as revealed by the differences between Fig. 10a, b.

A.3 Pump wavelength measurements for saturated absorption data

Simultaneous recordings of the 1083 nm pump wavelength and of saturated absorption signals from the ancillary cell (C_{lock} in Fig. 4) are reported here. Figure 11 displays saturated absorption data and fitting steps for a frequency scan over the C_{S} to D_{1} lines in an isotopic mixture cell. The experimental frequency offsets were inferred from the recorded wavelength values using our nominal frequency offset (\nu^{(4)}/\hbar in Table 2) and the wavelength in vacuum \lambda_{2} (Eq. A.1) for the D_{2} transition:

\nu = \nu^{(4)}/\hbar + c \{(1/\lambda - 1/\lambda_{2})\}.

(A.2)

For convenience data processing consisted of two steps: the demodulated signals (left) were fit using Gaussian profiles and the residues (right) were fit using Lorentzian profiles. The positions of the saturated absorption dips, which had typical FWHMs of 20 MHz, were thus precisely inferred.

Series of recordings comprising three back and forth scans were repeated on four different days using two gas mixture cells. The line position fits from each scan were combined to compute the three line splittings D_{1}−D_{2}, D_{2}−C_{S}, and C_{S}−C_{S} plotted in Fig. 12a−c. As was observed for the probe laser scans (Sect. 4.4), a significant scatter of results was found for these experiments.

Figure 12d–f displays the results of the same recordings, where the frequency offsets inferred from the wavemeter data were replaced with the laser frequency control voltage data in the horizontal axes of Fig. 11.
Fig. 12 Results of saturated absorption data yielding fit positions of the $C_8$, $D_2$, $C_9$, and $D_1$ lines for series of 3 repeated scans (see Fig. 11). Top row, frequency splittings (a–c). Bottom row, frequency control voltage differences (d–f). a, d for the $D_1$–$D_2$ interval. b, e for the $C_8$–$C_9$ interval. c, f for the $C_8$–$D_2$ interval. The solid and dashed lines in graphs (a–c) are the averaged data and computed values, respectively.

Table 8 Statistics for frequency splitting data compiled in Fig. 12. Columns 3 to 5 contain the differences between the averages of the fit frequencies and the computed values, the standard deviation and the standard error of the fits. The first three lines are data from wavemeter readings, the last three lines from control voltage values (see text, data types $\lambda$ or $V$). $N_s$ is the number of scans used in the averages.

| Line names | Data type | Av.-th. ($\text{MHz}$) | SD ($\text{MHz}$) | SE ($\text{MHz}$) | $N_s$ |
|------------|-----------|------------------------|------------------|------------------|------|
| $C_9$–$C_8$ | $\lambda$ | 33.7 | 135 | 28 | 23 |
| $D_2$–$C_8$ | $\lambda$ | 31.9 | 131 | 24 | 29 |
| $D_2$–$D_1$ | $\lambda$ | −51.5 | 61 | 11 | 23 |
| $C_9$–$C_8$ | $V$ | −15.9 | 20.7 | 4.2 | 23 |
| $D_2$–$C_8$ | $V$ | −15.8 | 16.1 | 3 | 29 |
| $D_2$–$D_1$ | $V$ | 11.7 | 15.5 | 2.9 | 23 |

The scaling factor between frequency and voltage differences was evaluated as the average of the ratios of the theoretical frequency splittings (the dashed lines in Fig. 12a–c) to the measured voltage splittings (in Fig. 12d–f).

The scatter of the voltage splittings was significantly lower than that inferred from wavelength data, as appears on the statistical summary of the plotted results listed in Table 8. A fourfold to eightfold reduction of the standard deviation of the three probed line splittings is evidenced, with a significant reduction of the differences from expected values (lines 4 to 6 vs. lines 1 to 3 in Table 8).

The results of these series of recordings are thus affected by the measurement errors of the wavemeter in a way similar to those of Sect. 4.4. Moreover, since the pump diode laser frequency is known to be reliably controlled by its temperature (therefore, by the con-
control voltage in these experiments), the large reduction of errors in line splittings expressed as functions of this voltage further demonstrates that the frequency dependence of the wavemeter errors was the limiting factor for the precision of our Doppler-free measurement on the 706.5 nm transition.

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