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Quantum Interference Effects in Saturated Absorption Spectroscopy of n=2 Triplet-Helium Fine Structure

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Over the past decade, precision measurements of the helium $2^3P$ fine-structure intervals have been performed using a variety of experimental techniques. We have shown in previous work (Phys. Rev. A 89, 043403, and references therein) that these measurements can be significantly shifted by quantum interference with neighboring transitions, despite the fact that the neighboring transitions are much broader than the uncertainty of the saturated-absorption signal. The interference shifts calculated in this work are larger than the uncertainty of the saturated-absorption signal. The interference shifts are taken into account.

The saturated-absorption method uses laser excitation in an rf-discharge helium cell, and detects the power of the laser beam after it has passed through the sample. The measurement is performed with and without a second counterpropagating laser beam. In both cases, only the forward-beam power is measured, and the interference between the two cases (i.e., with and without the counterpropagating laser beam) forms the saturated-absorption signal. Because of Doppler shifts, only when the laser is tuned to near resonance do both laser beams communicate to the same atoms, and therefore the saturated-absorption signal has a much narrower line width than the full Doppler-broadened laser resonance.

We calculate first the interference effects for the $2^3S_1,m_J=1—2^3P_1,m_J=1$ ($|1⟩—|2⟩$ in Fig. 1) saturated-absorption measurement. This measurement is shifted due to interference between this transition and the $2^3S_1,m_J=1—2^3P_2,m_J=1$ ($|1⟩—|3⟩$ in Fig. 1) transition. The measurement can be modeled using a closed four-level system, since any population that starts in the metastable $|1⟩$ state can only be excited by a linearly-polarized laser beam to $|2⟩$ or $|3⟩$, and these can radiatively decay only to $|1⟩$ or $|0⟩$. The $2^3S_1,m_J=0$ state ($|0⟩$) can be considered to be a dark state because the $2^3S_1,m_J=0—2^3P_1,m_J=0$ transition is electric-dipole-forbidden.

II. DENSITY-MATRIX EQUATIONS

To model the saturated-absorption technique for the measurement performed in [17], we consider a laser with frequency $\omega=2\pi f$ and linear polarization $\hat{z}$ interacting with a cloud of metastable helium atoms. The forward beam and the counterpropagating beam travel in the $+\hat{y}$ and $-\hat{y}$ directions, respectively, and both laser beams have a uniform intensity $I_0$ over the volume of atoms that contribute to the signal. Each Doppler group within the atomic cloud (with velocity $v_y$ in the direction of the

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laser-beam) is considered separately, with the full signal obtained by averaging over all Doppler groups. An atom with velocity component $v_y$ sees equal and opposite Doppler shifts $\pm \Delta \omega_D = \pm 2\pi \Delta f_D = \pm \omega v_y/c$ for the two laser beams. The total electric field experienced by a moving atom in the laser beams is $\vec{E}(t) = E(t) \hat{z}$, with

$$E(t) = E_0 \cos[(\omega + \Delta \omega_D)t] + sE_0 \cos[(\omega - \Delta \omega_D)t + \Delta \phi].$$

Here, $E_0 = \sqrt{\frac{\hbar}{\eta \omega_0}}$, $\Delta \phi$ is the phase difference between the two laser beams, and the parameter $s = 1$ or 0 indicates if the counterpropagating laser beam is switched on or off.

As was done in [21], one can write density-matrix equations describing the transfer of population in the four-level system of Fig. 1, where the population begins in the metastable $|1\rangle$ state, and the laser is nearly resonant with the $|1\rangle - |2\rangle$ transition. Since very little population is transferred to the far-off-resonance $|3\rangle$ state, the full set of equations can be approximated [21] by a reduced set of modified density-matrix equations involving only the initial state $|1\rangle$ and the on-resonant state $|2\rangle$:

$$\dot{\rho}_{11} = \left( i\Omega_2 \frac{2}{2} + \frac{i\Omega_3 \gamma_{23} - 1}{2\omega_{23}} \right) \rho_{12} + \gamma_{2 \rightarrow 1} \rho_{22},$$

$$\dot{\rho}_{12} = \left( i\Omega_2 \frac{2}{2} - \frac{i\Omega_3 \gamma_{23} + 1}{2\omega_{23}} \right) \rho_{12},$$

$$\dot{\rho}_{22} = \frac{i\Omega_2 \frac{2}{2} \rho_{12} - \frac{i\Omega_2 \frac{2}{2}}{\tau} \rho_{12} - \gamma_2 \rho_{22}.}$$

Here $\omega_{23}$ is the fine-structure splitting between $|2\rangle$ and $|3\rangle$ (see Fig. 1), while $\Delta_2 = 2\pi \Delta f$ is the detuning of the laser from the $|1\rangle - |2\rangle$ transition frequency. The radiative decay terms involve the quantities $\gamma_2 = \gamma$, $\gamma_{2 \rightarrow 1} = \gamma/2$, and $\gamma_{23} = -\gamma/2$, where $\gamma = 1/\tau$ and $\tau = 98$ ns is the lifetime of the $2^3P_J$ states.

We are using the rotating-wave approximation, and the Rabi frequencies $\Omega_2$ and $\Omega_3$ associated with the driven transitions between the metastable state $|1\rangle$ and the two excited states $|2\rangle$ and $|3\rangle$ are given by

$$\Omega_i(t) = \Omega_i^{(+)} + s\Omega_i^{(-)},$$

$$\Omega_i^{(+)} = e^{i\omega_D t} \frac{E_0}{\hbar}(1|z|i),$$

$$\Omega_i^{(-)} = e^{-i(\Delta \omega_D t + \Delta \phi)} \frac{E_0}{\hbar}(1|z|i).$$

The (+) and (−) superscripts refer to the forward and counterpropagating beams, and, as in Eq. 1, the value of $s$ indicates whether the counterpropagating beam is switched on or off.

For Eq. 2, the effect of state $|3\rangle$ is small since $\omega_{23} = 2\pi(2291$ MHz) is more than 1000 times larger than the other frequency scales ($\gamma$, $\Omega_i$, and $\Delta_2$, all of which are $\lesssim 1/\tau$). As in [21], a small parameter $\eta$ is defined as the ratio of these two frequency scales, and $\rho_{23}$ is of order $\eta^2$, and can be ignored. The order-$\eta$ density-matrix elements $\rho_{13}$ and $\rho_{23}$ are eliminated from the full set of equations, to give Eqs. 2(a)-(c), which are complete to first order in $\eta$. The order-$\eta$ terms in Eq. 2 that result from this elimination are: $\frac{\Omega_3^2}{\Delta_{23}}$, which is the usual ac-Stark shift of the $|1\rangle$ state, and $\frac{\Omega_3 \gamma_{23} - 1}{2\omega_{23}}$, which represent interference effects in the decay pathways.

### A. Absorption lineshapes

The saturated-absorption method measures the absorption of the forward laser beam ($\Omega_2^{(+)}$ of Eq. 3(b)). For an atom interacting with the laser for a time $T$ the total absorption is

$$\alpha_s = \frac{i}{2} \int_0^T \left( \Omega_2^{(+)} \rho_{12} - \Omega_2^{(+)} \rho_{12} \right) dt.$$
This integral, when combined with $\rho_{12}$ obtained by integrating Eq. 2, is complete to order $\eta$. All of the integrations are performed numerically, and must be repeated twice: for $s=0$ and $s=1$ (i.e., with and without the counterpropagating beam), as indicated by the subscript $s$ in Eq. 4.

The numerical calculation of the difference in absorption $\Delta \alpha=\alpha_0 - \alpha_1$ must be repeated for a range of frequencies near resonance to obtain a saturated-absorption lineshape. Such lineshapes must be calculated separately for each Doppler group ($\Delta \omega_D$ of Eq. 1), as well as for each relative phase ($\Delta \phi$ of Eq. 1) and the average, $\Delta \alpha$, gives the full lineshape, as shown, for example, in Fig. 2. Since the saturated-absorption lineshape is dominated by atoms that are nearly at rest, it is sufficient to average only over the values of $|\Delta \omega_D| < 80$ MHz.

Figure 3 shows the FWHM of the calculated saturated-absorption lineshape (solid lines) as a function of laser intensity $I$ and of interaction time $T$ (top axis). The feature is broadened both at short interaction times ($T<2\pi\tau$) and above saturation ($\Omega_2 T > 1$), but can approach the natural width (dashed line) for sufficiently long $T$ and low enough $I$.

Collisions of $n=2$ helium atoms in the discharge with the much larger number of ground-state atoms in the cell determine the interaction time $T$. Velocity-changing collisions move atoms to different Doppler groups, and therefore the rate of these collisions determines the time $T$ spent in the Doppler group that contributes to the saturated-absorption signal. Thus, as in [17], an approximate correspondence can be made between the reciprocal of the pressure $P$ in the cell and the interaction time $T$: $T=C/P$. To find the constant $C$, we plot measured [22] saturated-absorptions widths (points in Fig. 3) and adjust $C$ (and therefore the bottom axis of Fig. 3) to obtain the best match between the experimental points and our calculated widths for a laser intensity of $I=0.1$ mW/cm$^2$.

III. INTERFERENCE SHIFTS

The quantum-mechanical interference effect that is being considered in this work (which leads to the $\omega_{23}^{-1}$ terms in Eq. 2) causes shifted lineshapes. After numerically calculating the saturated-absorption lineshape, the half-maximum points (see Fig. 2) are used to determine the shift. As shown in Fig. 4, the shift depends on how many Doppler groups are included in the calculation, and is fully converged if all Doppler groups with $|\Delta f_D|<20$ MHz are included.
The analysis thus far has been for the $2^3S_1, m_J=1\rightarrow 2^3P_1, m_J=1$ transition. Identical shifts are obtained when considering the $m_J=-1$ transition. Also, a similar calculation can be carried out for a laser nearly resonant with the $2^3S_1\rightarrow 2^3P_2$ transitions. Every part of the analysis is similar, except for a sign which results from the fact that in this instance the on-resonance process is at a lower (rather than higher) frequency than the off-resonant ($2^3S_1\rightarrow 2^3P_1$) process. A concern that arises when considering the $2^3S_1\rightarrow 2^3P_2$ transition is that more than four states could be involved, since any population that decays to the $2^3S_1,m_J=0$ state (state $|0\rangle$ in Fig. 1) could be excited to the $2^3P_2,m_J=0$ state. However, as in [17], a sufficiently large magnetic field can remove the degeneracy of the $m_J$ levels, and therefore suppress this transition, leading once again to a four-level system.

In the saturated-absorption measurements in Ref. [17], the $2^3P_1\rightarrow 2^3P_2$ fine-structure interval is determined by subtracting the observed linecenters for the $2^3S_1\rightarrow 2^3P_1$ and $2^3S_1\rightarrow 2^3P_2$ intervals. Because of the equal and opposite shifts, the net shift in the fine-structure interval is two times larger than the shift for the individual transitions, and this shift is shown using solid curves in Fig. 5 for a range of interaction times $T$ and laser intensities $I$.

For the measurement in [17], the most precise data were taken for helium cell pressures between 18 and 38 mTorr, and were then extrapolated to zero pressure. The thick lines in Fig. 5 show extrapolations of our calculated shifts from this same pressure range. Note that this pressure range avoids the larger interference shifts calculated for lower pressures (larger $T$) and larger $I$ in Fig. 5. These larger shifts result from the larger line widths (see Fig. 3) for these $T$ and $I$, which allows for more interference with the neighboring resonance. With the extrapolations to $P=0$, the interference shifts still remain nonzero. This holds true even in the limit of zero laser intensity $I$. Thus, a repetition of experiments down to lower and lower pressures and laser intensities will not eliminate the interference shift. As indicated by the lowest-intensity thick dashed line, the interference shift in the limit of zero $I$ and $P$ is $-1.2$ kHz. This is larger than the 0.51-kHz uncertainty of the saturated-absorption measurement [17] of this interval, and thus it must be carefully considered and corrected for in these measurements.

The thin dashed lines in Fig. 5 show the shifts that result if the $\Omega_{323-323}$ term in Eq. 2 is artificially suppressed. These dashed curves therefore give only the shifts due to the $\Omega_{323-323}$ term (the ac-Stark shift term). This part of the shift does go to zero in the limit of zero $P$ and $I$, as indicated by the thick dotted lines, and therefore it is the $\Omega_{323-323}$ term which leads to the $-1.2$ kHz shift.

The net $-1.2$ kHz shift can be broken up into three parts. The shift at 18 mTorr and 0.1 mW/cm$^2$ (the pressure and intensity at which the most precise data were taken in [17], as indicated by the circle in Fig. 5) is $-1.6$ kHz. The extrapolation to zero pressure (the thick solid line) causes an additional shift of $-0.6$ kHz. Finally, an extrapolation of the $P=0$ intercepts of Fig. 5 to zero intensity causes an additional shift of $+1.0$ kHz.

The extrapolations of our calculated shifts to $P=0$ and $I=0$ are certainly not exact. For example, a helium cell at a pressure $P$ would have a range of times $T$ between velocity-changing collisions (ranging from approximately 50% to 150% of the average $T$), and therefore the correspondence between $T$ and $P$ assumed in Fig. 5 is only approximate. Furthermore, the current modeling does not explicitly include magnetic fields, and the degree to which state $|0\rangle$ of Fig. 1 is a dark state might depend on magnetic field, on laser intensity, and on the pressure in the cell. Because of such uncertainties, we include a 50% uncertainty for each of the components of the shift and estimate a total shift of $(-1.6 \pm 0.8$ kHz) $+(-0.6 \pm 0.3$ kHz) $+(+1.0 \pm 0.5$ kHz) $=-1.2\pm1.0$ kHz for this measurement.

Fig. 6 gives a summary of the current experimental (a - d) and theoretical (e) determinations of the $2^3P_1\rightarrow 2^3P_2$ fine-structure interval. In that figure, the solid error bars are measurements without corrections for interference effects. Each measurement uses a different technique (microwave separated oscillatory fields (a), laser spectroscopy in a laser-cooled beam (b), laser spectroscopy in a well-collimated thermal beam (c), and saturated-absorption spectroscopy in a cell (d), respectively), and therefore each has different interference processes, and requires a separate calculation for the interference shift.

The first three of these calculations have been previously completed ([20], [16], and [18]), and the correction for the fourth is considered in this work. The correction is small for the microwave measurement, both because it has a subnatural line width and because the separation...
between the observed resonance and its nearest neighbor is an order of magnitude larger than in the other cases. Even the sign of the shifts depends on the details of the measurement technique (as shown, e.g., in [19]), and thus it is coincidental that the shifts are similar for the other three measurements. The corrected measurements (which include the interference effects and their estimated uncertainties) are shown as dashed error bars in Fig. 6. Note that the corrected values show much better agreement with each other and with theory than the uncorrected values.

IV. CONCLUSIONS

We have calculated the effect of quantum-mechanical interference with a far-off-resonant transition on the saturated-absorption measurements of the $2\ ^3P_1$–$2\ ^3P_2$ fine-structure interval in atomic helium. The effect is found to cause a shift that is large compared to the measurement uncertainty, and therefore must be properly accounted for. The shift is significant even though the off-resonant transition is separated by 2.3 GHz, or 1400 natural widths. This work illustrates that shifts due to quantum-mechanical interference must be carefully considered for all precision measurements. When possible, measurements should be planned to minimize the interference effect (as, for example, for point a of Fig. 6), by either ensuring that the separation between resonances is very large, or by techniques which suppress any nonresonant processes. To correct for the interference shifts, a calculation must be performed based on the exact measurement technique used.

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