Limitations of rotating-wave approximation in magnetic resonance: characterization and elimination of the Bloch–Siegert shift in magneto-optics

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Abstract

We present investigations of radio-frequency (RF) resonances observed in an optically pumped rubidium vapor. By measuring the systematic shifts (the Bloch–Siegert shifts) of RF resonances in low magnetic fields, we demonstrate limitations of the rotating-wave approximation in the case of angular momentum $F \geq 1$. The resonance shifts and deformations are characterized in a wide range of parameters and it is shown that the observed behavior is far more complex than in a standard two-level system. It is also demonstrated that the shifts can be controllably turned on or off by switching between the oscillating and rotating magnetic field. Experimental results are supported with numerical calculations, reproducing all features of the observed signals. Besides fundamental aspect of the research, application of rotating magnetic field helps to suppress/evaluate spectroscopic-measurement and precise-metrology systematic errors. The reported study has also important implications for quantum metrology and information processing beyond RWA and standard two-state qubit dynamics.

1. Introduction

A standard treatment of a two-level system interacting with a resonant electromagnetic field is based on decomposition of an oscillating field into two counter-rotating components and consideration of system’s interaction with only one of them. This approach, known as rotating-wave approximation (RWA), is justified in most cases, such as not-too-strong or optical fields, i.e. when the field–atom coupling strength is a small fraction of bare frequency of an uncoupled atom. In a particular case of oscillating magnetic field $B_{RF} \cos(\omega t)$ applied to polarized atoms immersed in perpendicularly oriented static magnetic field $B_0$, the interaction leads to appearance of the resonance at frequency $\Omega_r$. In RWA, $\Omega_r$ equals the static-field Larmor frequency $\Omega_0 = g\mu_B B_0 / h$, where $g, \mu_B, h$ are the gyromagnetic ratio, the Bohr magneton, and the reduced Planck constant, respectively. However, due to interaction with a counter-rotating component of the oscillating magnetic field, the actual resonance position is shifted from $\Omega_0$. The lowest-order approximation of this shift was primarily derived by Bloch and Siegert [1] and is given by

$$\Delta\Omega = \Omega_0 - \Omega_r \approx \frac{\Omega_{RF}^2}{16\Omega_0},$$

where $\Omega_{RF} = g\mu_B B_{RF} / h$. $\Delta\Omega$ is known as the Bloch–Siegert shift (BSS).

Since its first description in 1940 [1], BSS was carefully investigated theoretically [2–4] to develop analytical solution for $\Delta\Omega$. Experimental studies of the effect were performed with different electron–spin [5, 6], as well as, nuclear–spin systems [7, 8]. Recently, the effect was revisited for novel applications in quantum metrology [9] and quantum information processing [10–12]. In the latter case, the interest is motivated by the need to perform coherent control of a quantum system with possibly short electromagnetic pulses to reach high bandwidth of...
system's manipulation. Since it is the pulse area that determines coherent evolution of the Bloch vector, shortening of the pulses must be done by corresponding increase of their amplitudes, which makes BSS more important. Consequently, for strong driving fields, when the shift becomes comparable with the eigenfrequency of the system (the Larmor frequency in our case), significant departures from RWA hinder coherent control of the qubits because of complex, nonharmonic Rabi oscillations. Recent work with superconducting circuits [10, 11] and nitrogen-vacancy centers in diamonds [13, 14] demonstrate that this difficulty can be successfully alleviated. The BSS appears important also for experiments with cold atoms in dressed and adiabatic potentials [15–17]. An interplay between the Bloch–Siegert and AC Stark effects has been studied and applied to control valleytronics properties of novel 2D materials like monolayer WS2 (see [18] and references therein).

Despite growing importance of BBS, to our knowledge, the effect has not been taken into consideration in studies of nonlinear magneto-optical effects in alkali metal vapors [19, 20] (although some estimates were given in [21]). Since these phenomena are widely used in modern quantum metrology, we find that thorough analysis of BSS is crucial to avoid systematic errors in precision magnetometry and spectroscopic measurements with warm and cold atoms [22, 23].

In this paper, we present both experimental and theoretical (numerical) studies of BSS in a Rb vapor system subjected to static and oscillating/rotating magnetic fields. The effect is studied by means of radio-frequency (RF) nonlinear magneto–optical rotation (NMOR) [24, 25], where resonances are created and detected via properly tuned linearly-polarized laser light interplaying with static and oscillating magnetic fields. NMOR is based on optical detection of medium’s anisotropy (birefringence and/or dichroism) caused by magnetic perturbations of optically polarized medium. The perturbation may change populations and coherences of the atomic system, so the method is useful for sensitive monitoring of atomic states and their superpositions [23, 26]. Important features of our system are: a possibility of application of either rotating or oscillating magnetic field under otherwise the same experimental conditions, generation of narrow (≈1 Hz) RF NMOR resonances, and measurements at ultra-low-oscillating- (RF-) field frequencies (ω/2π = 1–100 Hz). The first aspect is essential for elimination of systematic errors, while the second and third are relevant because of inverse proportionality of the shift to the magnitude of static (DC) magnetic field (equation (1)). Precision of our method, enhanced by the small value of Ω0, enables utilization of much weaker RF fields than in previous experiments. Hence, in our case the perturbative approach derived in [2–4] is valid. Exerting strong RF fields to reach the limit where perturbation method is no longer credible (i.e. ΩRF > 2.4Ω0) leads to broadening and deformation of resonance lineshapes, and thus reduced precision of the measurement [2, 4].

In RF NMOR, RWA is typically applied with respect to all oscillating fields (optical and magnetic) and BSS is disregarded. Herein, the second approximation is not used. Moreover, in contrast to most previous studies with spin 1/2, we analyze the case of angular momentum F > 1. In that case, a strong RF field (ΩRF ≫ Ω0) reveals important interference effect. The effect goes beyond the standard two-state qubit dynamics and enters the avenues to generate and control more sophisticated superposition states, like qutrits, quunits, etc [27].

2. Numerical approach

We investigate BSS of NMOR resonances using the density-matrix formalism and a model atomic system with the F = 1 ground state and the F′ = 0 excited state, interacting with linearly polarized light and immersed in AC and DC magnetic fields (figure 1). Despite its simplicity, this is a generic system for most NMOR experiments. The Hamiltonian of the system is presented in a rotating frame, corresponding to the optical frequency ωlight, which is the only approximation used in our numerical modeling. In the |F, mF⟩ base, with a quantization axis along the light propagation direction (z) but perpendicular to the static magnetic field, the Hamiltonian of the system under consideration is given by

$$H = H_0 + H_0 + H_B,$$

where $d$ is the electric dipole moment of the optical transition, $E_{\text{lin}}$ is the light vector of an amplitude, $\Delta = \omega - \omega_{\text{light}}$ is the laser frequency detuning, and $\beta = -dE_{\text{lin}}/\hbar$ is the optical Rabi frequency. For the the magnetic field composed of the static and oscillating magnetic field, $B_{\text{lin}} = \hbar/(\gamma I_p) [0, 0, \Omega_0, \Omega_{\text{RF}} \cos(\omega t)\frac{2}{T}]$, the magnetic Hamiltonian $H_B = -\mu \cdot B$, where $\mu$ is the magnetic dipole moment, takes the form
results in modulation of the rotation angle

Excited state.

As the indices are related to the optical coherence between specific sublevels, in our system, the polarization

rotation angle is given by

\[ \phi \propto \text{Im}(\rho_{-10} - \rho_{10}) \]

where \( \rho_{-10} \) and \( \rho_{10} \) are the optical coherence between the \( |1, -1\rangle \) (\( |1, 1\rangle \)) ground state and the \( |0, 0\rangle \) excited state. Time dependence of \( \mathbf{B}_{RF}(t) \) results in modulation of the rotation angle \( \phi \). To reproduce measurements of the

2 Numerical calculations were performed by means of Atomic Density Matrix package [29] in Wolfram Mathematica computing environment.
light parameters with the phase-sensitive detection, time-dependent solutions of the polarization rotation is are integrated over the period \( T = \frac{2\pi}{\omega} \) at the time \( t_S \) needed for the system to approach the steady state

\[
\phi_p(\omega) + i\phi_q(\omega) = \int_{t_S}^{t_S+T} \phi(\tau) \exp(i\omega\tau) \, d\tau,
\]

where subscripts \( p \) and \( q \) correspond to the signal measured in phase and in quadrature with the AC magnetic field.

3. Experimental setup

The central part of our experimental setup (figure 2(a)) is a spherical, paraffin-coated vapor cell of 10 cm diameter filled with isotopically enriched \(^{85}\)Rb (longitudinal relaxation time \( \approx 1 \) s) at room temperature. The cell is surrounded by a three-layer magnetic shielding made of \( \mu \)-metal and a ferrite cube as an additional innermost layer. The shielding is able to reduce residual magnetic fields by four orders of magnitude (from Earth’s magnetic field of \( \sim 1 \) G (\( 10^{-4} \) T) to \( \sim 100 \) \( \mu \)G (\( 10^{-8} \) T)) in a wide range of frequencies [30]. A set of magnetic-field coils generating homogeneous magnetic fields in all three directions is mounted inside the shield. The leading field is set along the \( y \) axis. Other coils serve for passive compensation of residual fields and are used to generate a variable magnetic field. In case of oscillating field, one of the perpendicular coils (\( x \) or \( z \)) is fed with a sinusoidal signal. The rotating field is produced when \( \cos(\omega t) \) and \( \sin(\omega t) \) signals are applied to \( x \) and \( z \) coils.

3 The cell construction and field homogeneity enable observation of narrow (\( \lesssim 1 \) Hz) resonances, which is essential for precise measurements. On the other hand, the efficient magnetic shielding and compensation make it possible to measure resonances at ultra-low frequencies (\( \lesssim 100 \) Hz), corresponding to DC magnetic fields of about 100 \( \mu \)G (\( 10^{-8} \) T) or smaller.
respectively. Due to imperfect symmetry between the coils some degree of ellipticity may be present in the rotating field. We mitigate this effect by optimizing rf field amplitude and phase in x coil (see below).

Linearly polarized laser beam tuned to the D1 (795 nm) line of rubidium creates spin alignment and monitors atomic polarization changes induced by magnetic fields. It is tuned to a wing of the Doppler broadened $F = 3 \rightarrow F'$ transition (detuned $-260$ MHz from the $F = 3 \rightarrow F' = 2$ transition). The laser light frequency is stabilized with a dichroic atomic vapor laser lock [31, 32] and a saturated absorption spectroscopy system serves as frequency reference. Typical light power$^4$ prior the cell is 20 $\mu$W (in paraffin-coated cells, the power is averaged over the cell volume [33]). Polarization rotation is detected by a balanced polarimeter. The signal is demodulated with a lock-in amplifier, operating at the RF field frequency typically between 1 and 100 Hz.

The RF NMOR signals obtained by sweeping the static magnetic field with a fixed RF-field frequency and amplitude are presented in figure 2(b). The Larmor frequency $\Omega_S$ corresponding to the static field is extrapolated from an independent calibration based on relation between the current applied to the calibrated coil and the resonance frequency in a high-frequency regime with low AC-field amplitude. The oscillating field generates two resonances at $\pm \Omega_S$, where the sign denotes the conventional precession and static magnetic field direction/handedness of RF field, whereas a single resonance at $\Omega_S$ occurs with the rotating field. In case of the latter, to ensure good quality of rotating field we adjusted the amplitude and phase of magnetic field in x coil to see minimum amplitude residual signal at $-\Omega_S$. In that manner we achieved <2% degree of ellipticity, measured as ratio between two signals’ amplitude. The in-phase component of the observed resonances is characterized by the dispersive shape, while its quadrature counterpart is absorptive. Width of both resonances is determined by the ground-state relaxation rate. A small dip around $\Omega_0 = 0$ is considered as an experimental singularity caused by 1/f noise in the system$^5$.

The drawback of the scheme with a fixed RF-field frequency is the necessity of scanning the static magnetic-field amplitude; due to the change in operation conditions of the current source, magnetic-field scanning may introduce uncertainties to the measurements (magnetic-field drifts). To avoid this problem, further results are taken with a constant static magnetic field (more stable operation conditions) and the RF-field frequency $\omega$ is swept discretely (point by point with 1 s intervals between successive measurements, related to lock-in integration time of 300 ms) around the resonance frequency $\omega_r$. The RF-field amplitude remains constant for each scan. A restriction of such a scheme is the ability of scanning only the positive frequencies of the RF field.

4. Results

Figure 3 presents numerical (derived from equations (9) and (10)) and experimental signals (in-phase component) generated with the oscillating and rotating fields at three distinct AC-field amplitudes (expressed in terms of the Rabi frequency $\Omega_{RF}$). At low $\Omega_{RF}$, the shape of the NMOR resonance is purely Lorentzian. When the AC field becomes stronger the resonance broadens and a new narrow structure develops in its center. This effect does not occur for standard two-level structures, where the signal broadens but remains a single Lorentzian. The so-called Majorana reversal is characteristic for systems with $F \geq 1$. It was observed and first described in [34], however its rigorous calculation was derived primarily by Majorana [35]. Consistent analytical results were obtained in [36], where the most general case of AC and DC magnetic fields orientation was studied and approximation methods were used. The effect was interpreted as the result of quantum interference between one- and three-photon processes, each contributing to the resonance with a different width and amplitude depending on $\Omega_{RF}$ [37]. The resonance lineshapes, including the Majorana reversal for RF NMOR, were calculated in [38] and we use them in our fitting procedure in the following form

$$
\phi_1(\omega) = \frac{\phi_0 \gamma^2 - 2\Omega_{RF}^2 + 4(\omega - \omega_r)^2}{\gamma^2 + \Omega_{RF}^2 + (\omega - \omega_r)^2},
$$

$$
\phi_2(\omega) = \frac{\phi_0 \gamma^2 + 2\Omega_{RF}^2 + 4(\omega - \omega_r)^2}{\gamma^2 + \Omega_{RF}^2 + (\omega - \omega_r)^2},
$$

where $\phi_0$ is the amplitude of the magneto-optical rotation.

$^4$ The light power should be sufficiently low to reduce possible power broadening of observed resonances.

$^5$ Modeling without the noise does yield signals without the central zero-field feature, but adding pink noise reproduces faithfully the signals of the figure 1 (a). It was also experimentally verified that the width of the central dip increases with the vapor temperature (which confirms thermal origin of the noise).

$^6$ $\Omega_{RF}$ was calibrated with a separate measurement. $\nu(t) = V_{dc} + V_{ac} \cos(\omega t)$ signal was applied to the z coil and the resonance frequency $\Omega_{RF} / (2\pi)$ was measured.
Typically uncompensated negative frequency perpendicular to the static magnetic resonances. This interpretation is consistent with our numerical simulations of resonances in tilted systematic algebraic shifts. Figure 3 shows that for stronger AC fields, small features at $\omega_r/2$ and $2\omega_r$ are observed in experimental signals. We attribute the features to the residual, not fully compensated, transverse magnetic data presented in equation (11) for three different AC field strengths indicated in right bottom corner of each graph. The static magnetic field $B_0$ was set in such a way that $\Omega_0/(2\pi) = 23$ Hz. Residual magnetic fields are responsible for appearance of weak resonances at $\omega_r/2$ and $2\omega_r$ observed in the experimental data for high $\Omega_0$. Statistical error bars are smaller than the graph marker.

The correct description of RF NMOR with oscillating magnetic field requires taking into account the counter-rotating component of linearly polarized light, which is associated with the second resonance at the negative frequency $-\Omega$, (not observed directly in the experiment). Omission of the resonance would lead to systematic algebraic shifts. Figure 3 shows that for stronger AC fields, small features at $\omega_r/2$ and $2\omega_r$ are observed in experimental signals. We attribute the features to the residual, not fully compensated, transverse magnetic fields, which may tilt the resulting $B_0$ direction. As a consequence, the quantization axis may no longer be perpendicular to the static magnetic field and this would produce additional small harmonic and subharmonic resonances. This interpretation is consistent with our numerical simulations of resonances in tilted fields. We fit those residual signals with small absorptive resonances with predefined positions ($\omega_r/2$ and $2\omega_r$) and a given width. The residual fields cause also a small mixing between two lock-in channels (in-phase and quadrature), which is taken into account by fitting the experimental data with a combination of the resonance shapes given by equations (11). In case of the rotating-field-induced resonance, residual fields are exclusively responsible for the mixing. When such a resonance is considered, an additional phase mixing is produced by counter-rotating component of the AC field (interference of the measured resonance with the tail of the resonance at $-\Omega$).

For quantitative measurements of the Bloch–Siegert effect, we focused on the difference between resonance frequencies when the driving field is either oscillating or rotating, $\Delta \omega = \omega_{\text{rot}} - \omega_{\text{lin}}$, and regarded this difference as the measure of BSS. In figure 3, the measured and simulated values of $\Delta \omega$ versus $\Omega_RF$ are presented for three different DC field strengths. Since the maximum value of the applied RF field, $\Omega_{RF} = 12$ Hz < $0.5\Omega_0/2\pi$, the analysis was limited to the lowest order of the perturbative expression [3, 4]. According to equation (1), the shift should be inversely proportional to the intensity of the static magnetic field and increase quadratically with the amplitude of the modulated magnetic field, These predictions are well confirmed with the data presented in figure 4. However, an anomaly of unknown origin is seen around $\Omega_{RF}/(2\pi) = 2.5$ Hz, where experimental BSS value seem to be amplified.

In figure 5, the resonance position is shown for both rotating and oscillating AC field as the function of $\Omega_{RF}$ ($\Omega_0/(2\pi) = 45$ Hz). Solid red (or grey) and dashed black lines represent numerical solutions while dots and squares are experimental datapoints. Experimental dependencies agree well with the theoretical curves. Particularly, the resonance position barely depends on the AC-field Rabi frequency for rotating-field-induced

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7 Typically uncompensated field is of order of 10 $\mu$G ($10^{-7}$T).
Figure 4. Results of (a) experiment and (b) numerical calculations performed for three different Larmor frequencies. Polynomial fits from numerical data are shown in experimental section (a) as solid lines. The shift depends quadratically (first-order approximation) on the RF Rabi frequency and scales inversely with the static magnetic-field strength. The negative value of the shift reflects conversion from the static magnetic field sweeping to the frequency sweeping of the AC field.

Figure 5. Resonance position as the function of $\Omega_{RF}$ for oscillating (black squares) and rotating magnetic fields (red or grey dots) at a static magnetic field of $\Omega_0/(2\pi) = 45$ Hz. Solid and dashed lines represent numerical solutions while squares and dots are experimental datapoints.
resonances, which illustrates efficient elimination of BSS. Experimentally measured BSS values appear slightly overestimated by the numerical results. A small systematic shift of the rotating-field results is most likely caused by the magnetic-coil asymmetry (non-zero oscillating component along the DC field) and correspondingly weaker RF modulation of the atomic polarization. Additionally, imbalance of two counter-rotating magnetic-field components in the rotating-field measurements, i.e. some degree of ellipticity, may lead to non-zero signal at \(-\Omega\), and hence algebraic shift of the resonance.

5. Conclusions

In conclusion, we have characterized the Bloch–Siegert shift (BSS) of magneto-optical resonances in a multilevel \((F \geq 1)\) system of Rb atoms \((F = 3 \rightarrow F' \text{ transition in } ^{85}\text{Rb})\) in a parameter range characteristic for precision measurements of nonlinear magneto-optical rotation. The most prominent effect is the evidence of a double resonance structure with features at \(-\Omega\), and \(+\Omega\), for the oscillating field and only one resonance \(+\Omega\), for the rotating field. Various features of the observed signals, such as the BSS, Majorana reversal, phase mixing, and additional resonances due to field inhomogeneity have been investigated both experimentally and theoretically. Taking into account systematic errors discussed in previous sections, as well as simplicity of our theoretical model, a reasonably good agreement with our numerical simulations based on the density-matrix calculations, was obtained.

This study improves our understanding of the processes of light interaction with energy-level structures with \(F > 1/2\). In addition to accurate characterization of the observed resonances, including their distortions, and demonstration of RWA limitations, a method not explored before in context of magneto-optical resonances in room-temperature alkali metal vapors was proposed to mitigate the BSS. Namely, the application of the rotating, rather than oscillating RF field with attenuated amplitude.

While our analysis is focused on magnetometric applications, this work presents a general novel platform to investigate energy-level structures richer than a simple two-level system (qubit) and may apply to many precision measurements beyond RWA with resonance techniques and multilevel systems. Specifically, the precision experiments aiming at searches for permanent electric dipole moment [39] and time-variation of fundamental constants [40] can benefit from the above analysis. Proper accounting for the discussed shifts and/or distortions of the resonance shapes will improve accuracy of NMR [41], quantum magnetometry and metrology. In field of coherent control and quantum information, the above analysis should facilitate state manipulation and information processing with strong fields and high transmission rates in atoms, solids, as well as, in circuit QED and new materials.

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