Dynamic Effects in Nonlinear Magneto-Optics of Atoms and Molecules

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(Dated: March 15, 2004)

A brief and selective review is given of dynamical processes arising in nonlinear interactions between light and resonant systems (atoms or molecules) in the presence of a magnetic field.

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OCIS codes: 020.0020, 270.1670.

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1. Introduction

The study of resonant nonlinear magneto-optical effects (NMOE), as well as the closely related electro-optical effects, is an active area of research with a rich history going back to the early work on optical pumping. These effects result from the resonant interaction of light with an atomic or molecular system, generally in the presence of additional external electromagnetic fields. A recent review by Budker et al.1 deals primarily with the atomic case; the molecular case is detailed in a monograph by Auzinsh and Ferber.2 In this paper, we focus on the dynamical aspects of such interactions, attempting to offer the reader a unified view on various diverse phenomena and techniques while avoiding excessive repetition of material already discussed in the previous reviews.

Nonlinear magneto-optical (coherence) effects are observed when optically induced polarization of a medium undergoes quantum beats under the influence of an external field, and so influences the polarization and/or intensity of a transmitted probe light beam. While this is inherently a dynamical process on the microscopic level, a macroscopic atomic or molecular ensemble quickly reaches a steady state if the external parameters are held constant in time. Thus, in order to observe the quantum beat dynamics, these parameters must be varied at a rate significant compared to the polarization relaxation rate. One approach, discussed in Sec. 3, is to produce polarization with a pulse of pump light, and then observe the effect of the subsequent quantum beat dynamics on the transmission of a weak probe beam. Alternatively, the method of beat resonances (Sec. 4) can be used, in which an experimental parameter [light amplitude (Sec. ??), frequency (Sec. ??), or polarization (Sec. 4C) or external field strength (Sec. 4D)] is modulated,
2. Quantum beats: collapse and revival

Quantum beats is the general term for the time-evolution of a coherent superposition of nondegenerate energy eigenstates at a “beat frequency” determined by the energy splittings. In this paper, we are primarily concerned with the evolution of a polarized atomic or molecular ensemble of given angular momentum that has had its Zeeman components split by an external field. For linear Zeeman splitting—the lowest-order effect due to a uniform magnetic field—the evolution is Larmor precession, i.e., rotation of the polarization about the magnetic field direction. For nonlinear splittings, such as the quadratic Stark shifts, the polarization evolution becomes more complex, and it can be useful to have multiple methods for characterizing the state of the ensemble at a given time.

As an example, we consider the nonlinear Zeeman shifts that occur when atoms with hyperfine structure are subjected to a magnetic field. For states with electron spin \( S = 1/2 \), such as in the alkalis, the splittings are given by the Breit-Rabi formula (see, for example, Ref. 3):

\[
\frac{\omega_M}{2\pi} = g_J \mu B + \Delta \left( 1 + \frac{4Mx}{2I+1} + x^2 \right)^{1/2},
\]

where \( x = (g_J - g_I) \mu B/\Delta \), \( g_J \) and \( g_I \) are the electronic and nuclear Landé factors, respectively, \( B \) is the magnetic field, \( \Delta \) is the hyperfine structure interval, and \( I \) is the nuclear spin.

Consider an atomic sample of cesium that is initially in a stretched state \( F_z = F \) with respect to the \( z \)-axis, subject to an \( \hat{z} \)-directed magnetic field. The perturbed energy eigenstates are the \(|FM_z\rangle \) eigenstates of the \( F_z \) operator. The stretched state along \( \hat{z} \) is a superposition of these nondegenerate eigenstates, and so quantum beats are seen in the evolution of the system.

The time evolution of each eigenstate is given by

\[
C_M \exp(-i\omega_M t)|FM_z\rangle
\]

where \( \omega_M \) is the angular frequency of the quantum beat. The plots one clearly sees that orientation present in the initial state collapses and revives in the process of the temporal evolution. Temporal variation of higher polarization moments give rise to higher-order-symmetry contributions to the probability surface (see also Fig. 2).

In order to explore the decomposition into polarization

\[1\] If the component \( |q| = \kappa \), this direction coincides with \( \hat{z} \) (Fig. ??). If, on the contrary, for a given \( \kappa \), \( q = 0 \), the angular-momentum spatial distributions corresponding to such multipoles is axially symmetric with respect to \( \hat{z} \). In this case, the symmetry axis of order \( \kappa \) is perpendicular to \( \hat{z} \).
moments further, it is useful to plot the norms of the polarization moments as a function of time (Fig. 2). We see that initially all possible moments are present, with the lowest-order moments predominating. At $t = \frac{\tau}{4}$ the even-order moments are at their maxima and the odd-orders are zero.

We can now connect these pictures of the atomic polarization state to an experimentally observable signal, e.g., the absorption of weak, circularly polarized light propagating along $\hat{z}$. To find the absorption coefficient, assuming that the upper-state hyperfine structure is not resolved, we transform to the $|J, M_J \rangle |I, M - M_J \rangle$ basis quantized along $\hat{z}$ and sum over the transition rates for the Zeeman sublevels. At short time scales (Fig. 3a) we see absorption modulated at the Larmor frequency, due to the precession of the atomic polarization about the $x$ axis. The absorption is minimal when the state is oriented along $\hat{z}$, and maximum when it is oriented along $-\hat{z}$. Looking at the envelope of this modulation at longer time scales, we see “collapse and revival” with period $\tau/2$ of the absorption oscillation amplitude (Fig. 3b). The maxima of the envelope are associated with the stretched states shown in Fig. 1 and the minima with the states having planar symmetry with respect to the $x - y$ plane. This can also be seen in Fig. 2, which shows that the absorption envelope is proportional to the norm of the $\kappa = 1$ (oriented) moment, and does not depend on the higher moments. While it is true in general that weak probe light is not coupled to atomic polarization moments of rank greater than two, the fact that the absorption is insensitive to the $\kappa = 2$ (aligned) moment is a consequence of our assumption that the upper state hyperfine structure is unresolved.
In Fig. 3c one can see the effect of the third-order shifts, which become important at longer time scales. The effect of these cubic nonlinearities is to reduce the contrast of the envelope function.

Collapse and revival phenomena similar to the effect described here have been observed in nuclear precession. In this work, spin precession of an $I = 3/2$ system, $^{201}$Hg, was studied and the slight deviations from linearity in the Zeeman shifts responsible for the collapse and revival beats were due to quadrupole-interaction shifts arising from the interaction of the atoms with the walls of a rectangular vapor cell.

**Additional references for this Section:**

3. Pulsed pump light: Photon echo

A conceptually straightforward technique for observing quantum beat dynamics is to

4. Beat resonances

A. **Amplitude resonances: synchronous optical pumping**

In a pulsed experiment such as described in Sec. 3 the beats will tend to wash out as the pulse rate is increased relative to the polarization relaxation rate. Atoms or molecules polarized during successive pulses will beat out of phase with each other, producing steady-state polarization if the quantum-beat frequency is slower than the relaxation rate, or destroying the macroscopic polarization of the medium entirely if the quantum-beat frequency is faster. However, time-dependent polarization is regained when the pulse rate is made equal to the quantum-beat frequency. The light pulses contribute coherently to the medium polarization, and the ensemble beats in unison. This phenomenon, exhibited when the pump light is amplitude modulated at a sub-harmonic of the quantum-beat frequency, is known as synchronous optical pumping or “optically driven spin precession”. It is particular case of a general class of phenomena known as **beat resonances**, which may also be exhibited when light frequency or polarization or external field strength is modulated, as discussed in later parts of this section.

Synchronous optical pumping has been used over the years in many applications. To give just one example, this method was employed in sensitive searches for a possible permanent electric-dipole moment of an atom ($^{199}$Hg) whose existence is only possible due to a violation of both parity and time-reversal invariance.

Note that parametric resonance can be detected also in the average (steady-state) transmission of the medium, rather than by detecting the synchronously modulated transmission.

B. **Frequency resonances: FM NMOR**

Frequency (rather than amplitude) modulation of the pump light can be used to produce an effect similar to that discussed in Sec. 4A. Here the optical pumping rate is modulated as a result of its frequency dependence. One example of this is nonlinear magneto-optical (Faraday) rotation with frequency-modulated light (FM NMOR). In this case, linearly polarized light near-resonant with an atomic transition propagates through a medium immersed in a magnetic field oriented along the direction of light propagation. The frequency of the light is modulated and, therefore, the rates of optical pumping and probing acquire a periodic time dependence. The lowest-order interaction with the linearly polarized light field creates ground-state atomic alignment via optical pumping, and the alignment rotates about the magnetic field with the Larmor frequency. Due to the symmetry of the aligned atomic state, the medium’s polarization and optical properties are modulated at $2\Omega_L$. Thus a resonance occurs when the modulation frequency $\Omega_m$ equals $2\Omega_L$: the atomic sample is pumped into a macroscopic aligned state which causes a periodic modulation of the plane of light polarization at the output of the medium. The amplitudes of various harmonics of the time-dependent optical rotation (where $\Omega_m$ is the reference frequency) can be measured with a phase-sensitive lock-in detector (Fig. 4). Additional resonances can be observed at higher harmonics (for which $n\Omega_m = 2\Omega_L$, where $n$ is the harmonic order).

Earlier studies of nonlinear magneto-optical rotation involved measurement of the optical rotation caused by the equilibrium atomic polarization resulting from the balance of Larmor precession with various mechanisms causing the atomic polarization to relax (e.g., spin-exchange collisions or wall collisions). Thus in these earlier studies, the magnitude of the optical rotation increased linearly with the applied magnetic field $B$ for $gF\mu_B \gamma_{rel}$ (where $\gamma_{rel}$ is the relevant rate of relaxation), but fell off for larger magnetic fields. In fact, such zero-field resonances are also observed in the magnetic field dependence of the in-phase FM NMOR signals (Fig. 4). For the zero-field resonances, $\Omega_m$ is much faster than both $\Omega_L$ and the optical pumping rate for the cell, so in this case the frequency modulation does not significantly affect the pumping process. On the other hand, as the laser frequency is scanned through the atomic resonance, there arises a time-dependent optical rotation, so the signal contains various harmonics of $\Omega_m$.

The technique of FM NMOR enables one to translate the narrow features in the magnetic-field dependence of optical rotation, normally centered around $B = 0$, to much larger magnetic fields. This is especially useful for increasing the dynamic range of NMOR-based magnetometers, for which the sensitivity to magnetic fields can reach $\sim 10^{-11} \text{ G}/\sqrt{\text{Hz}}$.

C. **Polarization resonances**
Fig. 4. Signals detected at the first harmonic (a,b) and second harmonic (c,d) of $\Omega_m$ as a function of longitudinal magnetic field. This experiment employed buffer-gas-free, paraffin-coated vapor cells containing isotopically enriched $^{87}$Rb. The laser was tuned near the $D_1$ line, laser power was $15 \, \mu W$, beam diameter $\sim 2$ mm, $\Omega_m = 2\pi \times 1$ kHz, and the modulation amplitude $\Delta \omega = 2\pi \times 220$ MHz. Traces (a,c) and (b,d) correspond to the in-phase and the quadrature outputs of the signals from the lock-in detector, respectively. The zero-field resonances observed in traces (a,c) are similar in nature to the usual resonances observed in earlier studies (see text). The quadrature components arise because of a phase difference between the "probe" modulation and the modulation of the optical properties of the atomic medium. (Aligned atoms produce maximum optical rotation when the alignment axis is at an angle of $\pi/4$ to the light polarization direction.) Figure is from the paper by Budker et al.\textsuperscript{20}.

D. Parametric resonances: Magnetic-field modulation

As mentioned in Sec. 4 A, one way to obtain beat resonances is to modulate the external field (e.g., the magnetic field), and consequently the Larmor precession frequency. This method was used by Dupont-Roc\textsuperscript{24} for sensitive atomic magnetometry, and is presently employed as a useful general nonlinear-spectroscopic technique (see, for example, Failache et al.\textsuperscript{25}) known as parametric resonance.

In a typical setup, linearly polarized resonant laser light (Fig. 5) traverses the atomic (or paramagnetic-molecular) medium, to which a magnetic field parallel to the light-propagation direction is applied. The magnetic field has two components—a nearly dc component (that can be slowly scanned) and an ac component with frequency much faster than the ground-state polarization-relaxation rate. Transmitted light intensity is monitored with a photodetector, the signal from which is analyzed with a lock-in amplifier referenced to the ac modulation of the magnetic field.

As in FM NMOR (Sec. 4 B, Fig. 4), there are two types of resonances that are seen when the dc magnetic field is scanned: the zero-field resonance (independent of the ac-modulation frequency) that only appears in the in-phase component of the signal, and the frequency-dependent resonances in both the in-phase and quadrature components. The former can be understood from the fact that, for resonant light, the transmission is a quadratic function of the applied magnetic field, while the latter arise because of a stroboscopic effect. As the ac field adds to or subtracts from the static field, it leads to acceleration or deceleration of the Larmor precession, respectively. Since Larmor precession acts to average out the polarization induced by cw pump light, the net optical pumping rate is dependent on the magnetic field strength. As a result, the optical pumping rate is modulated at the ac frequency, and so—as discussed in Sec. 4 A, a beat resonance occurs when the modulation frequency is a subharmonic of the quantum beat frequency. Thus the optical pumping rate starts to precess away from the axis of the light polarization, the precession rate increases and passes through a maximum (in the case of a resonance) when the alignment is perpendicular to the direction of light polarization, after which the precession rate slows down once again as the axis of the alignment approaches the polarization axis. Thus, the light interacts efficiently with atoms precessing with a “correct” phase, and we have conditions for creating a large overall atomic polarization, which is then detected in transmitted light.
tensity (as the transmission through an aligned medium depends on the relative direction of the linear light polarization and the alignment axis).

As we see, the principle of parametric resonance with magnetic-field modulation is quite similar to that of FM NMOR. However, we point out that the latter scheme, under specific experimental circumstances, may have advantages over the latter. For example, if one is working with paramagnetic atoms in dc fields $\sim G$ (the Earth magnetic field range), the resonance is achieved for modulation frequencies in the MHz-range. In order to achieve a large modulation index, one needs to have a field-modulation depth $\sim 1$ G. Such a modulation may be technically more difficult compared to modulating a diode-laser frequency with a MHz-rate with a large modulation index.

E. Relation to CPT

For completeness, it is also important to mention the intrinsic connection of synchronous optical pumping and beat resonances with $\Lambda$-resonances, coherent population trapping, and mode crossing (see Ref. 26 for a review), all of which were discovered much later, and are based on essentially the same physics involving synchronous excitation.

The frequency of Larmor precession of a polarized medium in a known magnetic field is a measure the magnetic moment of the medium; conversely, the applied magnetic field strength can be measured with atoms or molecules pumped in a state with a known gyromagnetic ratio. In 1961, Bell and Bloom\textsuperscript{13} first demonstrated the appearance of resonances in transmission of the light whose amplitude was modulated at frequencies close to the frequency of the Larmor precession. An interpretation of the effect in terms of synchronous optical pumping was discussed in Section ??.

A similar approach is applied in Section ?? devoted to FM NMOR and also in the recent papers on optical-pumping magnetometry with frequency modulated light\textsuperscript{27} where, similarly to the work of Bell and Bloom Bell and Bloom\textsuperscript{13}, resonances in light transmission are detected. An equivalent explanation can be given in terms of coherent population trapping (CPT) resonances, which allows one to build a bridge between to the recent work\textsuperscript{27} also devoted to atomic magnetometry with frequency modulated light.

Indeed, harmonic modulation of the light intensity leads to the appearance of two sidebands at frequencies shifted from the unperturbed light frequency $\omega_0$ by the value of the modulation frequency: $\omega_{1,2} = \omega_0 \pm \Omega_m$. With modulation depth less than 100%, the spectral component with frequency $\omega_0$ also survives - Fig. 6 a. A $\Lambda$-resonance occurs when a difference in frequencies between a pair of spectral components of the modulated lights coincides with the frequency corresponding to the energy separation between the lower-state Zeeman sublevels (Fig. 6 b), which is the basic scheme for the CPT effects\textsuperscript{26}. For the simplest case of a $F = 1 \rightarrow F' = 0$ transition (Fig. 6 b), in the conditions of a CPT resonance, the population of a “dark” state increases (which is a coherent superposition of the $M = \pm 1$-sublevels decoupled from the light), and light transmission increases. For the case of harmonic modulation of the light intensity, two resonances are observed\textsuperscript{13} for the two possible pairs of the frequency components with $\Delta\omega_{1,2} = \Omega_m, 2\Omega_m$.

Similarly to the case of the amplitude modulation, CPT resonances also occur in the case of frequency-modulated light. For example, reference Andreeva et al.\textsuperscript{27} considered application of the CPT effect with frequency modulated light to atomic magnetometry. In this case, the light spectrum consists of an infinite number of sidebands with amplitudes of the n-th sideband given by a Bessel function $J_n(m)$ corresponding to the modulation index $m$. With modulation index $m \approx 1.5$ used in Ref. Andreeva et al.\textsuperscript{27}, only a few sidebands up to $n = 4$ are prominent. In the experiment Andreeva et al.\textsuperscript{27}, Cs atoms in both vacuum and buffer-gas cells were used, and the minimum observed width of the CPT resonance was 1.4 kHz in the latter case. The width of the resonance sets the lower bound on the magnetic fields (and, correspondingly, the resonance frequency) where the resonances can be resolved.

The FM NMOR technique (Sections ?? and ??) that also use frequency-modulated light for optical pumping can also be described in the language of CPT resonances. In the experiments\textsuperscript{20–22}, Rb atoms were contained in a paraffin-coated cell with long ground-state relaxation time, so very narrow resonances with widths $\approx 2\pi \times 1$ Hz were obtained. One of the advantages of using narrow resonances (in addition to the high sensitivity) is that it is possible to work at low magnetic fields. In Refs.\textsuperscript{20–22}, the light frequency was modulated according to $\omega_m \cos(\Omega_m t)$ with $\omega_m \approx 30$ MHz and $\Omega_m \approx 100 – 1000$ Hz. Under these conditions, the modulation index is very large, $m = \omega_m/\Omega_m \approx 10^5$ (for a discussion of modulated light, see, for example,\textsuperscript{7} ). In this case, the pairs of $\Delta M = 2$ sublevels are coupled by a very large number of frequency-sideband pairs with comparable amplitude (Fig. 6 c), and the description in terms of the CPT-resonances becomes less intuitive than the synchronous-pumping picture presented in Section ??.

The recent intensification of the use of light-frequency modulation for synchronous optical pumping is related, in particular, to the development and broad use of single-mode diode-laser systems. For such lasers, frequency modulation via the diode current and/or the cavity length controlled with a piezoelectric transducer voltage
appears to be much simpler and more robust than light amplitude modulation. However, such frequency modulation is usually accompanied with inevitable intensity modulation of up to $\sim 15\%$\footnote{Such parasitic modulation and the laser-intensity noise limit the performance of the magnetometers with transmission detection, which is not the case for the NMOR-based magnetometers\cite{Cohen-Tannoudji0} which detect optical rotation.}

The above mentioned ability of the FM NMOR technique with ultra-narrow resonances to sensitively measure low magnetic fields was recently applied to detection of nano-Gauss-level fields produced by a nuclear magnetization of a laser-spin-exchange-polarized gaseous-xenon sample placed near a Rb vapor-cell of the atomic magnetometer\cite{Cohen-Tannoudji0}. (Detection of nuclear magnetization with atomic magnetometer was demonstrated by \textit{et al.} et. al.)\cite{Cohen-Tannoudji0} The new generation of atomic magnetometers may prove ideally suited for novel low-field and remote-detection nuclear magnetic resonance (NMR) and magnetic-resonance imaging (MRI) applications.

The investigation Yashchuk et al.\cite{Yashchuk21} of FM NMOR effects associated with the optically pumped high-rank atomic polarization moments opens promising possibilities for magnetometry application, allowing one to work at higher light power with better statistical sensitivity and without significant increase in polarization relaxation rate due to power broadening.

5. Applications

A. High-order polarization moments

The technique of FM NMOR provides a direct method to create and detect higher order atomic polarization moments.\cite{Yashchuk21} Atomic alignment (responsible for the effects discussed in the previous section) is described as a rank $\kappa = 2$ polarization moment (quadrupole). Multipole moments of rank $\kappa \leq 2$ can be easily generated and detected with weak light (since photons have spin one), but higher-rank moments require multiphoton interactions for both production and detection. \textit{Marcis says: I think that we should be careful here. If we observe laser induced fluorescence which is caused by a WEAK probe field it still directly reflects ground state multipole moments of rank up to 4, see discussion about molecules further in this section.}

When the light-atom interaction is modulated at the frequency $\Omega_m = \kappa \Omega_L$, a resonance in both pumping and probing can occur, due to the symmetry of atomic polarization with rank $\kappa$ (Fig. 7, see also Sec. 2). Consequently, for sufficiently intense light, high-rank polarization moments can be generated as atoms interact with the light field multiple times at periodic intervals before the polarization relaxes. For example, the rank $\kappa = 4$ hexadecapole moment requires at least two light-atom interactions in order to be created. The optical properties of the medium are then modulated at $\kappa \Omega_L$ leading to a time dependent optical rotation at the first harmonic of $\Omega_m$. The detection of the higher-order multipole moment also requires a multiphoton interaction. In the case of the hexadecapole moment, two light-atom interactions are required at the probe stage as well, so the amplitude of the FM NMOR signal due to the hexadecapole resonance scales as the fourth power of the light intensity, as experimentally observed by Yashchuk et al.\cite{Yashchuk21}

Figure 8 shows the magnetic field dependence of FM NMOR signals from a paraffin-coated cell containing $^{87}$Rb, where the atoms are pumped and probed with a single light beam tuned to the $D1$ transition. At relatively low light power [Fig. 8(a)], there are three prominent resonances: one at $B = 0$, and two corresponding to $2\Omega_L = \Omega_m$ (see the previous section, Fig. 4). Much smaller signals, whose relative amplitudes rapidly grow with light power, are seen at $4\Omega_L = \Omega_m$, the expected positions of the hexadecapole resonances.

Various experimental signatures of high-rank polarization moments were previously detected with other techniques\cite{Yashchuk21}, but by taking advantage of the unique periodicity of the dynamic optical signals using FM NMOR, such high-rank moments can be directly created, controlled, and studied. Because of the enhanced optical nonlinearities and different relaxation properties, higher-order polarization moments are of particular interest for application in many areas of quantum and nonlinear optics.

Now let us assume that, due to a nonlinear light interaction with a molecular ensemble, high-rank multipole moments are created in the ground state. If we use a weak probe light to observe the ground-state molecules, this probe light connects the ground-state multipole moments to the excited-state ones. For example, excited state population (the rank-zero moment) will depend on the ground state population, orientation and alignment. Excited state alignment will be directly connected by a dipole transition to all existing ground state moments from population ($\kappa = 0$) to the hexadecapole moment ($\kappa = 4$). Generally, a dipole transition can

![Figure 7. Angular-momentum probability surfaces that visually represent various polarization moments: (a) “pure” quadrupole $\kappa = 2, q = 0$, (b) $\kappa = 4, q = 0$ hexadecapole, (c) same as in (b), but rotated by $\pi/2$ around the x-axis, (d) the average of (b) and (c), which has a 4-fold symmetry with respect to rotations around $\hat{x}$. In all cases, the minimum necessary amount of $\rho_0^{(0)}$ was added to ensure that all sublevel populations are non-negative. Figure from Ref. 21.](image-url)
connect ground-state multipole moments to the excited state ones with ranks satisfying $|\kappa_{gr} - \kappa_{exx}| \leq 2$. At the same time, it is known that the three excited state multipole moments – population, orientation and alignment directly influence fluorescence intensity and polarization in a dipole transition. Thus, if after a strong pump pulse which creates different high-rank multipoles in the ground state of molecules, we monitor the ground-state transient processes by observing fluorescence induced by weak probe light, then ground-state multipole moments of rank up to 4 can be observed directly. Such ground-state relaxation kinetics after ground-state depletion with a laser pulse was observed for K$_2$, molecules for multipole moments of up to hexadecapole.

If, in addition to the pulsed ground-state depletion, an external magnetic field is applied, then quantum beats with up to four times the Larmor frequency can be observed in the kinetics of laser-induced fluorescence. Such effects were also studied with potassium dimers. It can also be noted that high-rank multipoles are responsible for certain specific features (such as additional narrow structures) of the nonlinear ground-state Hanle-effect signals with continuous excitation.

Effects of high-rank multipoles in molecules can also be observed with harmonically modulated laser excitation. In Section ?? we discussed beat resonances in ground-state molecules. Because ground-state multipole moments of rank $\kappa \leq 4$ can be observed in fluorescence excited by a probe beam, one can observe beat-resonance signals at frequencies up to four times the Larmor frequency. Such a resonance occurring at the laser frequency equal four times the Larmor frequency was observed for the first time in the ground state of K$_2$ by Auzinsh and Ferber (Fig. 9). Subsequently, this resonance was studied in more detail.

Fig. 9. Ground-state beat-resonance signal measured as a change of the degree of polarization of laser induced fluorescence as a function of modulation frequency of excitation light. The experiment was done with K$_2$ molecules. Two resonances, at twice (alignment) and four times (hexadecapole moment) the Larmor frequency, were observed. Figure is from Ref. 37. Let us change the figure, to replace “double” and “quadrupole” with “twice” and “four times”, respectively!

B. Magnetometry

6. Polarization-noise spectroscopy, quantum nondemolition measurements, spin-squeezing

Consider an unpolarized ensemble of atomic spins in a static magnetic field. Suppose that the orientation of the ensemble is probed with weak light propagating perpendicular to the magnetic field (for example, by measuring time-dependent optical rotation). It turns out that, while no average orientation occurs for any observation frequency, the spectrum of the orientation noise displays a resonance at the Larmor frequency. The origin of this resonance can be understood by once again going into the rotating frame. At any given time, for $N$ atoms within the volume of the light beam and for any given spatial direction, in general, there is an imbalance on the order of $\sqrt{N}$ between the number of atoms oriented along and opposite to this direction. A characteristic time for
the persistence of the imbalance is \(1/\gamma\). Thus, measuring magnetization in the rotating frame, one would detect noise spectrum peaked at zero frequency with a characteristic width of \(\gamma\). Now, going back to the laboratory frame, this peak is shifted to the Larmor frequency, as observed experimentally. This experiment was done with a Na vapor cell with buffer gas and a focused dye-laser light beam with wavelength tuned near the 589-nm resonance line.

In the above discussion, it was sufficient to assume that the angular momentum (and associated magnetic moment) of each atom is represented by a classical vector. This analysis can also be carried through in a quantum-mechanical approach. An interesting consequence of the uncertainty relation between Cartesian components of the angular momentum is that noise similar to that in the experiment of is present not only for unpolarized samples, but also even for fully polarized samples with appropriate experimental geometry. (For example, the noise would still be present in the same experimental geometry if the atoms were fully polarized along the magnetic field and would have infinite longitudinal relaxation time. The width of the noise resonance is given by the inverse transverse relaxation time: \(\gamma = 1/T_2\).)

Dear co-authors: please read the following stream of consciousness carefully. Let us now briefly touch upon some fundamental quantum-mechanical issues of noise and measurement as relates to magneto-optics. According to general principles of quantum mechanics, a measurement perturbs the quantum state of the system under observation. For example, a photon can be absorbed from the probe light-beam exciting the atom up from the state one is attempting to measure. It is important to realize, however, that if one is not attempting to extract the complete information about the system, it is quite straightforward to set up a quantum nondemolition (QND) measurement that will not alter the quantity one is trying to determine [see, for example]. Specifically, in our example where orientation in a given direction is measured via optical rotation of the probe beam, note that photon-absorption probability scales with detuning from optical resonance \(\Delta\) as \(1/\Delta^2\), while optical rotation due to the imbalance of the number of atoms oriented along and opposite to the light-propagation direction scales as \(1/\Delta\). Thus, a QND measurement of orientation is realized by simply tuning the light sufficiently far away from resonance. Dear co-authors: I am convinced that the above does indeed capture the main idea behind QND. However, I am a bit confused about something. If we can measure population non-perturbatively, one would think we should be screwing up the phase. However, it does not seem to me that any phase gets screwed up here, except in a a well-characterized way, due to ac Stark.

Marcis: This sounds very interesting. I will think about this more carefully.