Part I

Principles and techniques

1

General principles and characteristics of optical magnetometers

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1.1 Introduction

Optical magnetometry encompasses the wide range of experimental techniques in which light is used to measure the response of atomic angular momentum to magnetic fields. Atomic magnetic moments $\boldsymbol{\mu}$ arise due to the magnetic moments associated with the intrinsic spins of constituent electrons and nuclei as well as electronic orbital motion. In the presence of a magnetic field $\mathbf{B}$, a torque

$$\boldsymbol{\tau} = \boldsymbol{\mu} \times \mathbf{B}$$

acts on the atoms. For sufficiently small magnetic fields the atomic magnetic moment $\boldsymbol{\mu}$ can be assumed to be independent of $\mathbf{B}$, in which case $\boldsymbol{\tau}$ causes the component of angular momentum transverse to $\mathbf{B}$ to precess about $\mathbf{B}$ at the Larmor frequency $\Omega_L = \gamma B$, where γ is the gyromagnetic ratio of the atomic species. When light propagates through and interacts with the atomic medium, angular momentum is exchanged between the atoms and the light field. Thus the angular momentum state of the atoms affects the angular momentum state of the light. In this way, precession of atomic angular momentum can be observed via the induced changes in the polarization and intensity of light interacting with the atoms, allowing optical measurement of Larmor precession.

The exchange of angular momentum between the atomic medium and light field that enables optical detection of Larmor precession can similarly be used to polarize the atomic medium through the process of *optical pumping* (see the comprehensive reviews [1]–[3]). Optical pumping refers to the process whereby photons absorbed by atoms transfer their angular momentum to the atoms, thereby creating polarization in both ground and excited states. Ground-state spin polarization is achieved through both depopulation of selected Zeeman sublevels and repopulation of Zeeman sublevels by spontaneous transitions from

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the excited states polarized by the incident light. In most optical magnetometers, the signals used to detect Larmor precession scale with the ground-state spin polarization, and consequently, optical pumping dramatically enhances magnetometer sensitivity.

Optical magnetometers, presently unmatched in both absolute accuracy and magnetometric sensitivity [4, 5], are universally based on these aforementioned principles: (1) optical pumping of atomic spin polarization, (2) time-evolution of atomic spin polarization due to the torque exerted on atomic magnetic moments by the magnetic field, and (3) optical detection of the evolved atomic spin polarization state through the effect of the polarized atoms on light propagating through the atomic medium. Within this basic framework, there is a remarkable diversity of experimental techniques and physical effects, many of which are explored throughout this book, in the cited references, and in online supporting material (available at www.cambridge.org/9781107010352), and undoubtedly still others that remain to be discovered.

1.1.1 Fundamental sensitivity limits

The fundamental quantum-mechanical uncertainty in the measurement of atomic spin projection constrains the potential sensitivity of optical magnetometers. The *spin-projection-noise-limited* (or *atomic shot-noise-limited*) sensitivity δB_{SNL} of a polarized atomic sample to magnetic fields[1] is determined by the total number of atoms N and the spin-relaxation rate Γ_{rel} for measurement times $\tau \gg {\Gamma_{\mathrm{rel}}}^{-1}$ [6]:

$$\delta B_{\mathrm{SNL}} \approx \frac{1}{\gamma}\sqrt{\frac{\Gamma_{\mathrm{rel}}}{N\tau}}\,. \tag{1.1}$$

Equation (1.1) can be understood by noting that a measurement of a single atomic spin for a time $1/\Gamma_{\mathrm{rel}}$ determines the Larmor precession angle with an uncertainty on the order of 1 radian. If the measurement is performed with N atoms, the uncertainty is reduced by $\sqrt{N}$, and if the measurement is repeated multiple times, the uncertainty is reduced by the square root of the number of measurements, which most efficiently is $\sqrt{\Gamma_{\mathrm{rel}}\tau}$.

As can be seen from Eq. (1.1), to achieve the highest possible precision in magnetometric measurements, it is advantageous to have the longest possible relaxation time for the atomic polarization, i.e., the smallest possible Γ_{rel}, as well as the largest possible N. Therefore, optical magnetometers typically are based on measurements of long-lived ground-state spin polarization [an exception is the ^{4}He magnetometer (Chapter 10), in which polarization of the metastable $2\,^3S_1$ state is used]. A variety of techniques can be employed to minimize Γ_{rel}: antirelaxation coating the walls of the vapor cell containing the atoms to reduce spin-depolarizing wall collisions [7–10] (see Chapter 11), filling the vapor cell with buffer gas to slow diffusion of atoms to the cell walls (see, for example, Refs. [11–15]), and even atom trapping and cooling [16, 17] (Chapter 9). Also of note are optical magnetometers

[1] Here we ignore factors of order unity that depend on the details of the atomic system and optical magnetometer scheme, such as the total atomic angular momentum and relative contributions of different Zeeman sublevels.

using particular condensed matter systems, such as nitrogen-vacancy centers in diamonds (Chapter 8) and alkali atoms trapped in condensed (superfluid or solid) helium [18–20], that have small $\Gamma_{\rm rel}$.

There is also a contribution to optical-magnetometer noise from the quantum uncertainty of measurements of light properties (photon shot noise). Optical detection of atomic spin precession is usually performed by measuring either the intensity or polarization of light transmitted through the atomic sample. There are certain intrinsic advantages to measuring the light polarization, in particular, reduced sensitivity to noise due to laser-intensity fluctuations. If, for example, atomic spin precession is detected by measuring optical rotation of the plane of transmitted light polarization [21], the photon-shot-noise-limited sensitivity to the optical rotation angle φ is

$$\delta\varphi \approx \frac{1}{2}\sqrt{\frac{1}{\Phi\tau}}\,, \tag{1.2}$$

where Φ is the probed photon flux (photons/s) detected after the atomic sample and $\delta\varphi$ is measured in rad$/\sqrt{\rm Hz}$. It should also be noted that light–atom coupling via AC Stark shifts can generate additional noise (see Ref. [22] and Sec 1.4.3). In optimal operation, the contribution of photon shot noise to overall magnetometric noise does not exceed the contribution from atomic spin-projection noise [6, 23]. Upon optimization of the atomic density n for a given volume V of the sample, where $N = nV$, for an atomic-vapor-based optical magnetometer the dominant spin-relaxation mechanism becomes either spin-exchange or spin-destruction collisions (depending on the details of the magnetometry scheme), in which case $\Gamma_{\rm rel} = \xi n$, and the optimum magnetometric sensitivity becomes

$$\delta B_{\rm opt} \approx \frac{1}{\gamma}\sqrt{\frac{\xi}{V\tau}}\,. \tag{1.3}$$

The relaxation constant ξ ranges between $\sim 10^{-9}$ cm^3/s and $\sim 10^{-13}$ cm^3/s for alkali atoms, depending on the details of the collisions [1]. Thus for optical magnetometers using alkali vapors, the optimal magnetometric sensitivity for a $V = 1$ cm^3 magnetic sensor ranges between 10^{-11} and 10^{-13} G$/\sqrt{\rm Hz}$ (1 to 0.01 fT$/\sqrt{\rm Hz}$). Issues related to noise in optical magnetometers, including exploration of squeezed states and quantum nondemolition (QND) measurements, are addressed in detail in Chapters 2 and 3.

1.1.2 Zeeman shifts and atomic spin precession

The language of atomic spectroscopy provides a complementary description of optical magnetometry: the light field propagating through the atomic medium measures the Zeeman shifts of atomic states. The Hamiltonian describing the Zeeman shift is

$$H_Z = -\boldsymbol{\mu}\cdot\mathbf{B}\,. \tag{1.4}$$

For an elementary particle such as the electron, the magnetic moment is directly proportional to the intrinsic spin $\mathbf{S}$, $\boldsymbol{\mu} = g_e \mu_B \mathbf{S}$, where g_e is the electron's Landé g-factor and μ_B is the Bohr magneton,

$$\mu_B = \frac{e\hbar}{2mc} = \frac{\alpha}{2} e a_0 \approx (2\pi\hbar) \times 1.4\ \text{MHz/G}\,, \tag{1.5}$$

where e is the absolute value of the electron charge, $\hbar$ is Planck's constant, m is the electron mass, c is the speed of light, $\alpha = e^2/(\hbar c)$ is the fine structure constant, and $a_0 = \hbar^2/(me^2)$ is the Bohr radius. The Landé g-factor for the electron, $g_e \approx 2.002\,32$, can be determined from extraordinarily precise experimental measurements [24–26] and from the theory of quantum electrodynamics [27], and thus, measurements of the Zeeman shifts of electron spin states constitute absolute measurements of the magnetic field.

For an atom the situation becomes more complicated. For sufficiently small magnitudes of $\mathbf{B}$, the atomic magnetic moment $\boldsymbol{\mu}$ is determined by internal atomic interactions and is approximately independent of $\mathbf{B}$,

$$\boldsymbol{\mu} = g_F \mu_B \mathbf{F}\,, \tag{1.6}$$

where $\mathbf{F} = \mathbf{I} + \mathbf{J}$ is the total atomic angular momentum ($\mathbf{I}$ is the nuclear spin, and $\mathbf{J} = \mathbf{L} + \mathbf{S}$ is the electronic angular momentum, $\mathbf{L}$ is the orbital angular momentum, and $\mathbf{S}$ is the electron spin) and g_F is the Landé factor corresponding to a state with a particular value of F. It is evident from symmetry that $\boldsymbol{\mu}$ is parallel to the total atomic angular momentum $\mathbf{F}$ (this follows, for example, from the Wigner–Eckart theorem – see the discussion in Refs. [28] and [29]). However, for larger magnitudes of $\mathbf{B}$ where the Zeeman shifts become significant compared to energy splittings caused by hyperfine and fine interactions, the magnetic moment $\boldsymbol{\mu}$ becomes dependent on the external magnetic field $\mathbf{B}$ due to mixing of atomic states (this is the nonlinear Zeeman effect, see Fig. 1.1 and the detailed discussion in Ref. [29]). Ultimately, then, the energy splitting ΔE between Zeeman sublevels in an atom, and consequently, the frequency Ω associated with time-evolution of the atomic angular momentum, is described by an expansion in powers of B:

$$\Delta E = \hbar\Omega = \gamma_0 + \gamma_1 B + \gamma_2 B^2 + \gamma_3 B^3 + \cdots = \sum_{n=0}^{\infty} \gamma_n B^n\,, \tag{1.7}$$

where the expansion coefficients γ_n depend on the particular atomic states and Zeeman sublevels involved. As seen in the lower plot of Fig. 1.1, for alkali atoms subjected to magnetic fields in the geophysical range of $\sim$0.5 G, there are nonlinearities of the Zeeman shifts with magnitudes of tens of hertz, often larger than the magnetic resonance linewidths, that must be accounted for in sensitive optical magnetometry schemes.

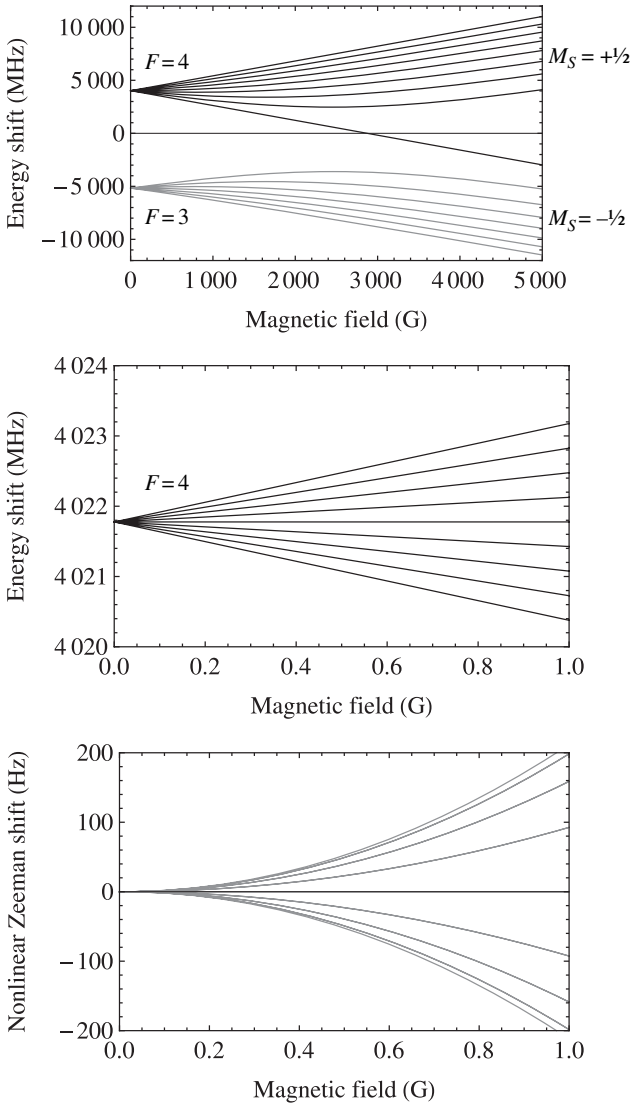

Figure 1.1 Top and middle plots: energies of the ground-state 6s $^2S_{1/2}$ hyperfine manifold of cesium ($I = 7/2$) as a function of applied magnetic field (the Breit–Rabi diagram). At low fields (middle plot), the atomic states are well described by the coupled basis where F and M_F are good quantum numbers and the Hamiltonian (Eq. 1.4) representing the interaction with the applied magnetic field is treated as a perturbation. At high fields, the atomic states are well described by the uncoupled basis where M_I and $M_J = M_S$ are good quantum numbers and the hyperfine interaction is treated as a perturbation. Bottom plot: nonlinear Zeeman shifts for the Cs ground state, computed by subtracting the linear term describing the low-field Zeeman shifts from the exact Breit–Rabi solutions.

1.1.3 Quantum beats and dynamic range

Since under all laboratory conditions Ω is very small compared to optical frequencies, and in many cases Ω is small compared to optical transition linewidths, Zeeman shift measurements typically involve observation of *quantum beats* [30–33]: the time-evolution of a coherent superposition of nondegenerate energy eigenstates at a frequency determined by the energy splittings. If a coherent superposition is created between two quantum states separated in energy by ΔE, according to the time-dependent Schrödinger equation, the complex phase between the states will evolve according to $\varphi(t) = \Delta E t/\hbar$, or in the case of Zeeman shifts, as described by Eq. (1.7), $\varphi(t) = m\Omega t$ where m is an integer. The time-evolution of $\varphi(t)$ causes the complex index of refraction for light propagating though the atomic medium to evolve in time as well, which can be observed in the time-dependence of the intensity and polarization of the light field.

If all fields, magnetic and optical, applied to the atoms are constant or vary in time slowly compared to the ground-state spin-relaxation rate $\Gamma_{\rm rel}$, the atomic spin polarization reaches a steady state. For spin-precession frequencies $\Omega \lesssim \Gamma_{\rm rel}$, atomic spins are unable to undergo an entire quantum-beat cycle before relaxing. In this case, although spin precession angles accrued between pump and probe interactions are random, because the angles are small compared to π the polarization does not average to zero. This produces a residual, magnetic field-dependent steady-state spin polarization that can be optically probed. For spin-precession frequencies $\Omega \gg \Gamma_{\rm rel}$, transverse atomic spin polarization is averaged out because probed atomic spins have precessed by random angles greater that π with respect to one another. In other words, atoms polarized at different times will, in general, beat out of phase with each other, canceling out the overall atomic spin polarization. For this reason optical magnetometers with constant or slowly varying fields have dynamic range limited by $\Gamma_{\rm rel}$ and find application as near-zero-field magnetometers.

Optical magnetometers used to measure fields corresponding to $\Omega \gg \Gamma_{\rm rel}$ therefore generally employ some type of time-dependent field: magnetic field pulses to induce transient responses, radiofrequency (RF) fields to take advantage of magnetic-resonance techniques, or modulated light fields to synchronously optically pump the atomic medium. For example, synchronous optical pumping (or "optically driven spin precession" [34]) creates time-dependent macroscopic atomic spin polarization for high quantum-beat frequencies ($\Omega \gg \Gamma_{\rm rel}$) by modulating the light at the quantum-beat frequency Ω or a subharmonic thereof. Polarization is produced in phase with that of atoms pumped on previous cycles, each optical pumping cycle contributing coherently to the atomic spin polarization, and as a result the ensemble beats in unison.

1.2 Model of an optical magnetometer

The basic principles of optical magnetometry are illustrated by considering an ensemble of two-level atoms with total angular momentum $F = 1/2$ in the ground state and $F' = 1/2$ in the upper state. Suppose that the atoms are immersed in a uniform magnetic field $\mathbf{B} = B\hat{\mathbf{z}}$

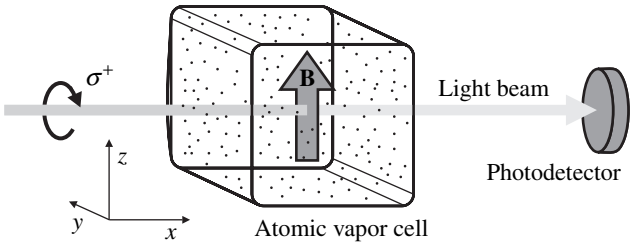

Figure 1.2 Example of a basic experimental setup for optical magnetometry.

and left-circularly polarized (σ^+) light,[2] near resonant with the (allowed electric dipole) $F \to F'$ transition, propagates along $\hat{\mathbf{x}}$ (Fig. 1.2). The strength of the light–atom interaction is parameterized by the *Rabi frequency*

$$\hbar\Omega_R = d\,\mathcal{E}_0\,, \tag{1.8}$$

where d is the transition dipole moment [for typical allowed electric-dipole transitions, $d \sim ea_0 \approx 2\pi \times 1.28$ MHz/(V/cm)] and $\mathcal{E}_0$ is the amplitude of the optical electric field. Spontaneous emission from the state F' back to F occurs at a rate Γ_0 (assume a closed transition). The optical pumping rate Γ_{pump} is given by Fermi's golden rule [28]:

$$\Gamma_{\text{pump}} = \frac{\Omega_R^2}{\Gamma_0}\,. \tag{1.9}$$

Let us assume that $\Gamma_{\text{rel}} \ll \Gamma_0$, $\Omega_R \ll \Gamma_0$, and ignore Doppler broadening (assume the atoms are stationary). Furthermore, to simplify our discussion, let us assume that $\Gamma_{\text{rel}} \ll \Gamma_{\text{pump}}$ (but note that for optimized magnetometer performance, typically $\Gamma_{\text{rel}} \gtrsim \Gamma_{\text{pump}}$). Under these conditions, the *saturation parameter* [28, 29]:

$$\mathcal{K} = \frac{\text{excitation rate}}{\text{relaxation rate}} = \frac{\Gamma_{\text{pump}}}{\Gamma_{\text{rel}}} = \frac{\Omega_R^2}{\Gamma_0\Gamma_{\text{rel}}} \gg 1\,, \tag{1.10}$$

indicates that the system is in the regime where the populations of ground-state Zeeman sublevels are strongly perturbed by the light field. Since $\Omega_R \ll \Gamma_0$, we can neglect stimulated emission and associated effects.

Suppose that initially $B = 0$ [Fig. 1.3(a)]. Interaction of the atoms with the light field causes ground-state optical pumping: choosing the quantization axis along the light propagation direction $\hat{\mathbf{x}}$, we see that the population of the $M_x = -1/2$ state (denoted $|-\rangle_x$) is depleted relative to the population of the $M_x = +1/2$ state (denoted $|+\rangle_x$), since due to angular-momentum selection rules the light field only causes transitions between the

[2] Here we use the spectroscopists' convention for left and right circular polarization, where a σ^+ photon (one with positive *helicity*, with the photon spin along its direction of propagation) is said to be left-circularly polarized, and a σ^- photon is said to be right-circularly polarized.

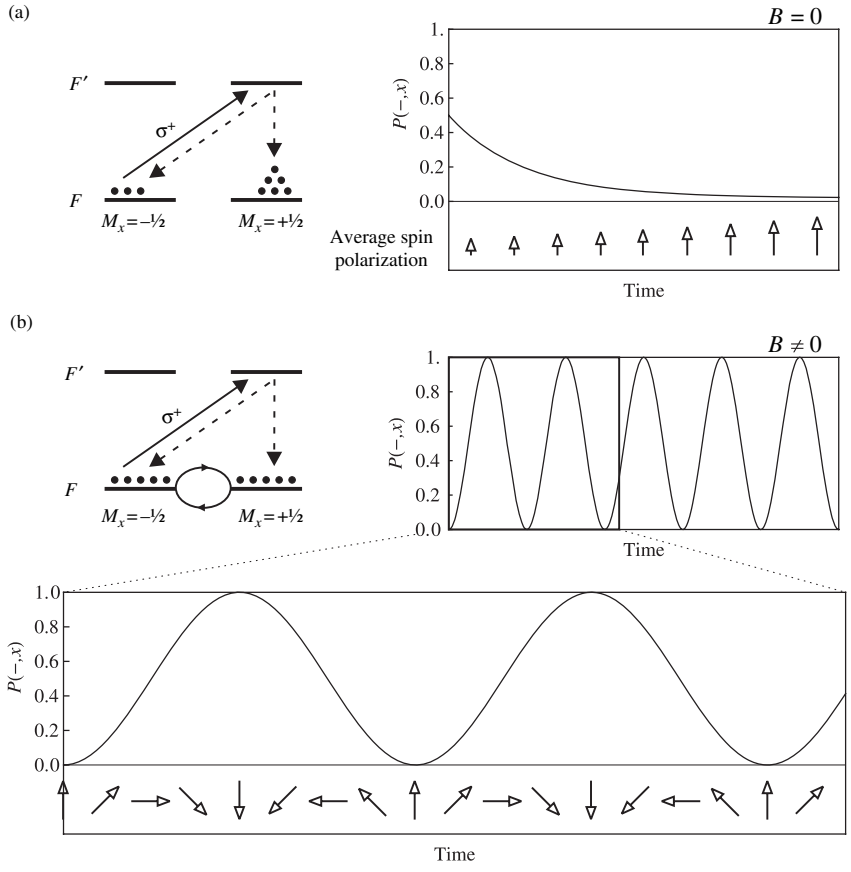

Figure 1.3 (a) Ground-state optical pumping of an ensemble of atoms with $F = 1/2$ in the ground state and $F' = 1/2$ in the upper state for the setup shown in Fig. 1.2 with $\mathbf{B} = 0$. Left-circularly polarized (σ^+) light excites the $M_x = -1/2 \rightarrow M'_x = +1/2$ transition, depleting the population of the $M_x = -1/2$ state. This decreases the probability $P(-,x)$ for atoms to be in the $M_x = -1/2$ state and increases the average spin polarization along x, as indicated by the increasing magnitude of the arrows along the bottom of the plot on the right. (b) Optical detection of spin precession with $\mathbf{B} \neq 0$. Larmor precession causes coherent oscillation between the $M_x = -1/2$ and $M_x = +1/2$ states (quantum beats) while the light probes the population of the $M_x = -1/2$ state [proportional to $P(-,x)$]. The coherent oscillation can also be understood as precession of the average spin polarization at the Larmor frequency, as shown by the arrows at the bottom of the lower plot.

$F = 1/2$, $M_x = -1/2$ and $F' = 1/2$, $M'_x = +1/2$ states. The atoms are optically pumped into a pure state $|+\rangle_x$ (since we can neglect ground-state spin relaxation under our assumptions) that does not interact with the light field. States that do not interact with light of particular polarization and/or frequency are commonly known as *dark states*. The atomic medium has acquired net angular momentum along $\hat{\mathbf{x}}$ from the light field.

Suppose that after optically pumping the atomic ensemble into $|+\rangle_x$, the magnetic field is suddenly (nonadiabatically) switched "on" ($B \neq 0$) and the light intensity is simultaneously