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**Resonant Magneto-Optical Rotation:
New Twists in an Old Plot**

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Dedicated to the centenary of the Macaluso-Corbino effect

Resonant Magneto-Optical Rotation: New Twists in an Old Plot

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Abstract

Nonlinear in laser power, near-resonant light-atom interaction leads to a number of spectacular modifications of a well-known effect – polarization plane rotation in a magnetic field applied along the direction of light propagation. In particular, the rotation can be enhanced by up to nine orders of magnitude compared to the linear case.

Faraday effect [¹] is the rotation of the plane of linear light polarization by a medium in a magnetic field applied in the direction of light propagation. In 1898, Macaluso and Corbino [²], studying absorption spectra of the alkali atoms in the presence of magnetic fields, discovered that the Faraday effect in the vicinity of resonance absorption lines also had a distinct resonant character.

The essence of the Macaluso-Corbino effect can be illustrated by considering a simple case of a $F=1 \rightarrow F'=0$ transition (Fig. 1). Linearly polarized light incident on the sample can be resolved into two counter-rotating circular components (σ^{\pm}). In the absence of a magnetic field, the $M=\pm 1$ sublevels are degenerate and the optical resonance

frequencies for σ^+ and σ^- coincide. When a magnetic field is applied, however, the Zeeman shifts lead to a difference between the resonance frequencies for the two circular polarizations. The refractive index dependences on the light frequency (the dispersion curves) are sketched in Fig. 2. A characteristic width of these curves Γ corresponds to the spectral width of an absorption line, which for typical experimental conditions in a vapor cell is dominated by the Doppler width and is on the order of 1 GHz for optical transitions. The difference between n_+ and n_- is shown in the lower part of Fig. 2. It leads to a difference in phase velocities of the two circular components of light and, as a result, to polarization plane rotation. In general, in addition to the difference in refractive index for the two circular polarizations (circular birefringence), there is also a difference in absorption (circular dichroism). Thus, a linear light polarization before the sample generally evolves into an elliptical polarization after the sample. In the ongoing, we will assume an optically thin sample, so that both optical rotation (defined as the angle between the initial polarization direction and the principal axis of the final polarization ellipse) and the ellipticity are both small and can be treated independently.

For nearly-monochromatic light (i.e. light with spectral width much smaller than the transition width), and for zero detuning, the optical rotation in the sample can be estimated as

$$\phi \sim \frac{\frac{g\mu B}{\Gamma/2}}{1 + \left(\frac{g\mu B}{\Gamma/2}\right)^2} \cdot \frac{l}{l_0} \quad (1)$$

Here l is the sample optical length, and l_0 is the absorption length. In this estimate we used the fact that near an absorption line, the amplitude of each dispersion curve (Fig.2)

is comparable to the resonance value of the imaginary part of the complex refractive index responsible for absorption. According to Equation 1, optical rotation is linear with B at small values of the field. If we keep light at zero detuning but continue increasing B , the rotation reaches a maximum at $g\mu B \sim \Gamma/2$ at a level ~ 1 radian per absorption length, after which it decreases, falling as B^{-1} in the limit of large fields (Fig. 3).

This elementary discussion essentially summarizes the essence of the resonant Faraday rotation in the linear case. The starting point for nonlinear Faraday rotation is the realization that the small-field optical rotation is inversely proportional to Γ (Equation 1), and that using the methods of nonlinear (in laser power) spectroscopy, it is possible to effectively reduce Γ to a much smaller width.

The simplest nonlinear effect leading to enhanced Faraday rotation is spectral hole burning [³]*. This is illustrated in Fig. 4. A narrow-band laser light with frequency tuned close to the center of a Doppler-broadened line interacts only with a particular velocity group of atoms. Due to velocity-selective optical pumping, there appears a so-called Bennett hole in the atomic velocity distribution (upper trace in Fig. 4). The width of this hole corresponds to the natural linewidth of the transition, typically ~ 1 -10 MHz. The effect of this narrow hole in the much broader Doppler profile on the Faraday rotation can be probed e.g. by a weak probe laser light tuned across the resonance. The Faraday rotation produced by atoms with such velocity distribution can be thought of as rotation produced by the Doppler distribution without the hole minus the rotation that would have been produced by atoms that were pumped out. This is illustrated on the lower trace of Fig.4. Because the hole width is much narrower than the Doppler width,

* In this brief paper, we do not attempt to provide a full bibliography on this vast subject. More complete references can, however, be found in the cited articles.

we have a correspondingly larger small field rotation (see Equation 1), which is also of opposite sign compared to the linear effect. In many experiments, instead of using separate pump and probe laser beams, one beam is used both to produce the hole and to probe the optical rotation. This corresponds to always probing the peak of the nonlinear effect. Thus, when the laser frequency is tuned across the resonance, the optical rotation exhibits broad (Doppler) spectral profile which looks like an absorption curve and has non-zero integral rotation, and not the characteristic Macaluso-Corbino profile with zero integral rotation shown in Fig. 2.

The next nonlinear effect that gives even higher small-field optical rotation is the coherence effect [^{4,3}]. Consider again atoms with total angular momentum $F=1$ which are not aligned initially. These atoms can be viewed as an incoherent mixture of the following states: $|M=0\rangle, \frac{1}{\sqrt{2}}(|M=1\rangle \pm |M=-1\rangle)$. Now consider laser light with frequency corresponding to an $F=1 \rightarrow F'=0$ transition. The first of these states can be excited to the $F'=0$ state only by z -polarized radiation (z -absorbing state), while it is decoupled from x and y -polarized light. Similarly, the other two states (which are coherent superpositions of the $|M = \pm 1\rangle$ Zeeman sublevels) are y and x -absorbing states, respectively. Suppose the laser light is polarized in the y direction. Optical pumping by this light leads to depletion of the y -absorbing state. If the corresponding saturation parameter is sufficiently high, the medium becomes transparent for the y -polarized radiation, leaving atoms in a superposition of the x and z absorbing states. This process is known as coherent population trapping [⁵] because as a result of optical pumping by linearly-polarized light (see Fig. 1), atoms in the $M=\pm 1$ substates are not completely

pumped out as would seem to be the case at first glance, but largely remain in a “dark” coherent superposition.

Atoms in this dark state do not interact with the light of the same polarization as pump light. However they can still absorb and refract light of an orthogonal polarization. This is, in effect, atomic alignment. An ensemble of aligned atoms constitutes a medium with linear dichroism and birefringence. The birefringence turns out not to be important for the coherence effect because the refractive index is unity on resonance. In the presence of a magnetic field, the atomic alignment axis created by the coherent population trapping precesses around the direction of the field with the Larmor frequency. The effect of this precession on the light polarization can be understood if one thinks of the atomic medium as a thin layer of polarizing material (e.g. like a Polaroid film, see Fig. 5). It is easy to see that rotation of the “polarizer” around the z-axis results in a linear polarization of light at the output of the sample which is rotated by an angle proportional to the optical density of the sample and to $\sin(2\theta)$, where θ is the angle between the transmission axis of the “polarizer” and the direction of light polarization. In order to describe optical rotation of cw laser light, one has to sum the effect of elementary “polarizers” produced at times $-\infty < t < 0$ and to include relaxation.

For sufficiently low light power and magnetic field, the rotation due to the coherence effect is once again described by Equation 1, but now the relevant relaxation rate Γ is that for the ground state alignment. In the absence of atomic collisions, this relaxation is determined by the transit of atoms through the laser beam (i.e. the aligned atoms fly out of the laser beam, while they are being replaced by “fresh” atoms from the

volume of the cell). For a few-mm diameter laser beam and for atomic velocities in a typical experiment, $\Gamma \sim 100$ kHz.

The two mechanisms of the nonlinear Faraday effect discussed above, the hole-burning and the coherence effect, produce narrow features (resonances) in the magnetic field dependence of the rotation (c.f. Fig. 3) with widths corresponding to their respective relaxation rates [3,6]. Below (Figs. 6,7), we will illustrate various contributions to the nonlinear Faraday effect using the data obtained in a recent experiment at Berkeley [7].

Even though the coherence effect gives a small-field rotation enhancement of some four orders of magnitude compared to the linear case, a considerable further enhancement is possible. One straightforward way to achieve this is to increase the diameter of the laser beam. Another approach, which is most suitable for experiments with dense atomic beams is to use separated pump and probe laser beams. This was independently suggested in [8] and [9], and experimentally realized in the latter work. Using the separated laser field method and a hypersonic atomic beam [10], it is possible to achieve a relaxation rate $\Gamma \sim 1$ kHz (determined by the atoms' transit time between the pump and the probe region), while maintaining $ll_0 \sim 1$ for the probe beam (with $ll_0 \gg 1$ for the pump beam).

Another elegant method to decrease the effective alignment relaxation rate is to use cells with an anti-relaxation coating on the inner walls. With an appropriate coating, it takes many alkali atom collisions with the wall to destroy atomic alignment. Therefore, an atom aligned by the laser beam can bounce around the cell for a long time maintaining the Zeeman coherence. Application of such cells to the nonlinear Faraday effect was first demonstrated by S. I. Kanorsky, A. Weis and J. Skalla [11]. They used a paraffin-coated

glass cell (18 mm diam. \times 18 mm long) containing cesium vapor and observed an optical rotation resonance with a width corresponding to an effective relaxation rate $\Gamma \sim 1$ kHz.

We have performed experiments with rubidium vapor in a paraffin-coated spherical cell (8 cm diam.). (This particular cell was manufactured in 1964). In order to create a well-characterized small magnetic field, the cell was surrounded by a four-layer magnetic shield, with a system of magnetic coils installed within the innermost shield allowing one to create a magnetic field with x, y and z components from -2 to 2 Gs. An external-cavity diode laser was used. Its frequency was tuned to the rubidium D2 ($^2S_{1/2} \rightarrow ^2P_{3/2}$) resonance at 780 nm. The Doppler-limited absorption profile on this line consists of four peaks corresponding to two ground-state hyperfine components for each of the two stable rubidium isotopes: ^{85}Rb (nuclear spin $I=5/2$, relative abundance 72 %) and ^{87}Rb ($I=3/2$, 28 %); the upper state hyperfine structure is unresolved. The laser could be tuned to any of the four components of the line. The vapor cell was at room temperature at which $l/l_0 \sim 5 \cdot 10^{-2}$ on the strongest line (^{85}Rb , $F=3 \rightarrow F'$).

Fig. 6 shows the magnetic field dependence of the Faraday rotation for the laser tuned near the peak of the ^{85}Rb ($F=3 \rightarrow F'$) resonance. The broadest structure in the Figure (clipped due to the limited magnetic field range) corresponds to the hole-burning effect. A characteristic width of this structure is ~ 10 Gs. Note that even at the extremes of the available magnetic field scan range (-2 to 2 Gs), the contribution of the linear Faraday rotation is some three orders of magnitude smaller than that of the nonlinear effect. The next narrower structure in Fig. 6 is due to the coherence effect. Its characteristic width ~ 100 mGs is determined by the atoms' transit through the laser beam. The experimental magnetic field dependence of the Faraday rotation is fit with a theoretical lineshape that

consists of several terms of the form of Equation 1 with different widths and amplitudes corresponding to the different contributions to the optical rotation. The fitting curve is shown in the figure along with the experimental data.

The coherence effect related to the preservation of atomic alignment during wall collisions manifests itself as the sharpest feature at near-zero magnetic fields. A magnetic field scan corresponding to a 2000 \times zoom (compared to Fig. 6) of this low-field region is shown in Fig. 7. The remarkably narrow width of this resonance in the magnetic field dependence of the rotation is 40 μ Gs (peak-to-peak), corresponding to $\Gamma \approx 56$ Hz. To our knowledge, this is the narrowest resonance of this type observed so far.

The ultimately small relaxation rates ($\Gamma \sim 1$ Hz for a ~ 10 cm diam. cell) and a corresponding enhancement (of about nine orders of magnitude compared to the linear case) of the nonlinear optical rotation can be achieved if one uses the best demonstrated coatings [¹²] that can support several thousand wall-collisions before an alkali atom loses Zeeman coherence. We are planning to realize this at Berkeley in the near future.

So far, we have only discussed optical rotation in longitudinal magnetic fields (i.e. magnetic fields applied along the direction of light propagation). An example of the optical rotation resonance lineshape in the presence of a constant transverse magnetic field is shown in Fig. 8. In order to achieve a quantitative description of lineshapes in arbitrary fields, we have used a generalization of a simple theoretical model developed in [¹³]. In this model, the optical rotation originates from a three-step process: 1. Atomic alignment by the incident linearly polarized light; 2. Free precession of the alignment in the arbitrarily directed magnetic field; 3. The effect of the atoms on light according to the rotating “polarizer” picture discussed above. A lineshape fit obtained with this model is

shown in Fig. 8 along with the experimental data. It is interesting to note how well the details of the lineshape are reproduced in spite of the naiveté of the model.

The nonlinear optical rotation can be applied to sensitive magnetometry (for both longitudinal and transverse fields). As a first application, we are using this method for measurements of residual magnetic fields and shielding factors in an advanced multi-layer magnetic-shielding system [7]. Another intriguing possibility is to replace the magnetic field in a nonlinear Faraday rotation experiment with an electric field [14]. This way, one could perform a sensitive search for linear Stark shifts in an atom, which could arise due to a violation of both parity and time-reversal invariance [15].

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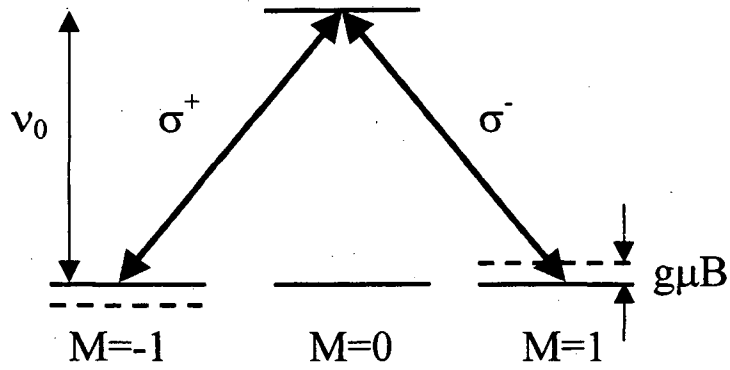


Fig. 1. An $F=1 \rightarrow F'=0$ atomic transition. In the presence of a longitudinal magnetic field, the Zeeman sublevels of the ground state are shifted in energy by $g\mu B \cdot M$. This leads to a difference in resonance frequencies for right- and left-circularly polarized light (σ^\pm).

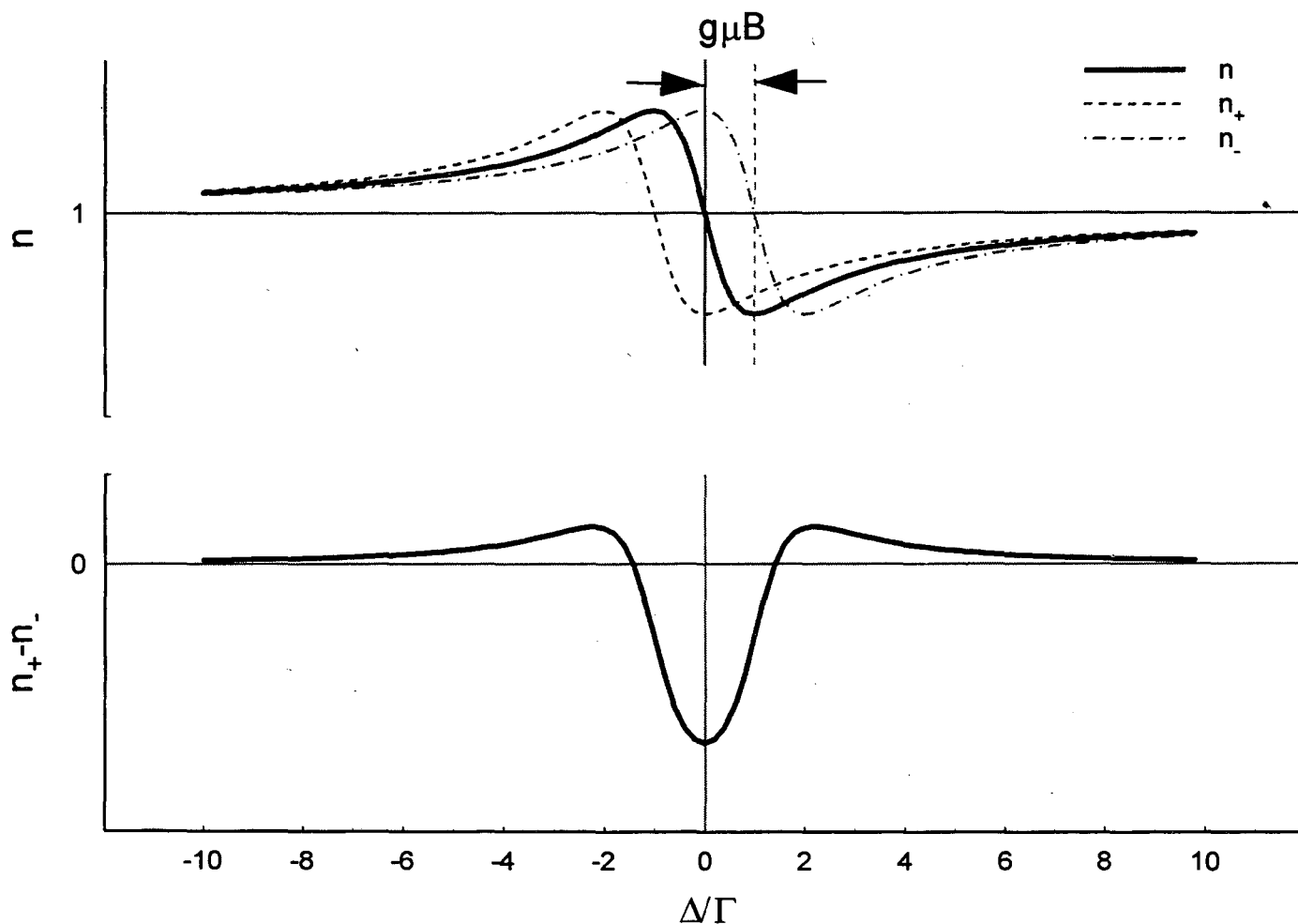


Fig. 2. The dependence of the refractive index on light frequency detuning Δ in the absence (n) and in the presence (n_{\pm}) of a magnetic field. Shown is the case of $g\mu B = \Gamma$ and a Lorentzian model for line broadening. Gaussian and Voigt models (see e.g. W. Demtröder. Laser Spectroscopy. Springer, 1996.), which are most appropriate in the case of a Doppler-broadened line, lead to qualitatively similar pictures. The lower curve shows the difference in refractive index for the two circular polarization components. This is the characteristic spectral profile of Macaluso-Corbino optical rotation.

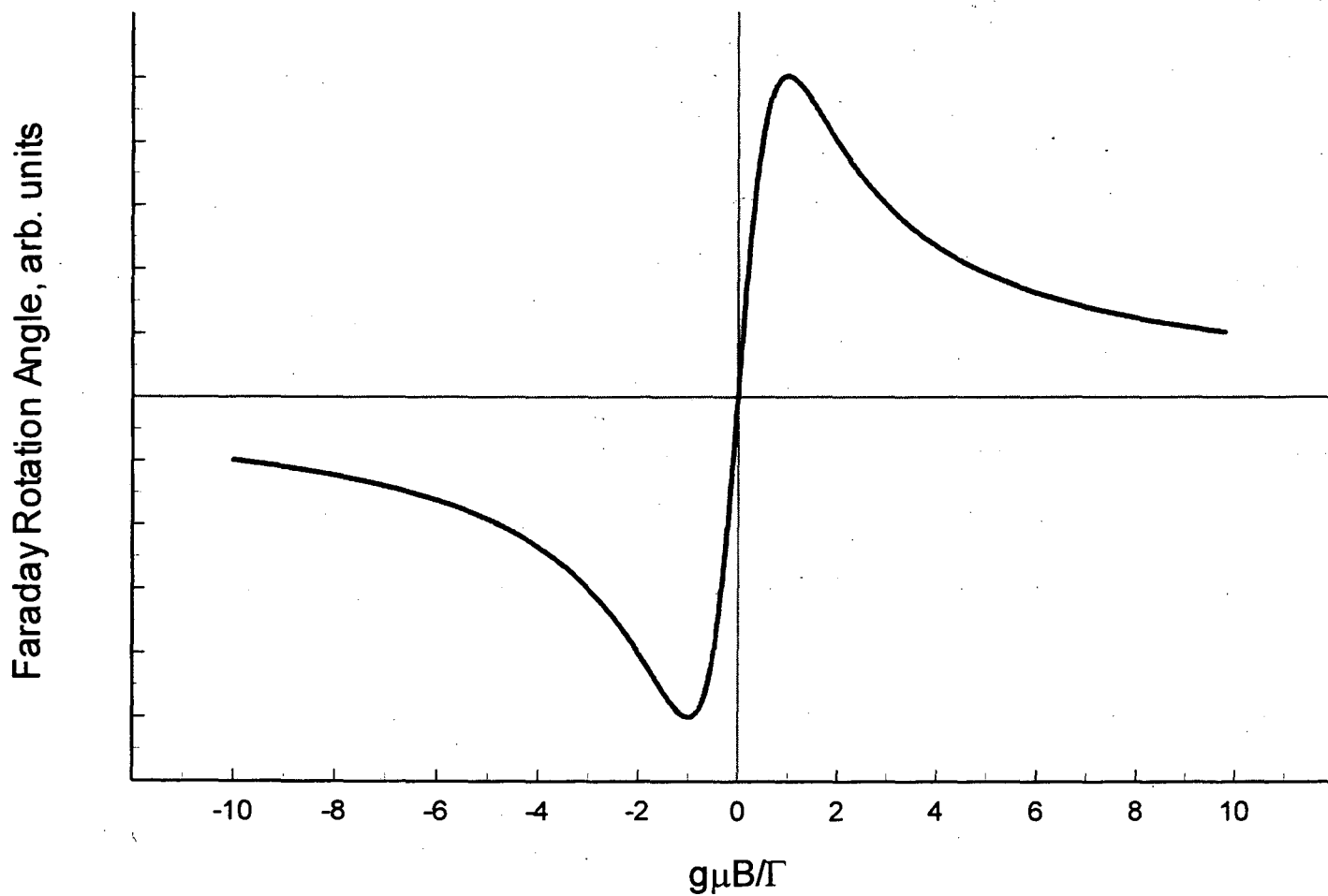


Fig. 3. Magnetic field dependence of the Faraday rotation.

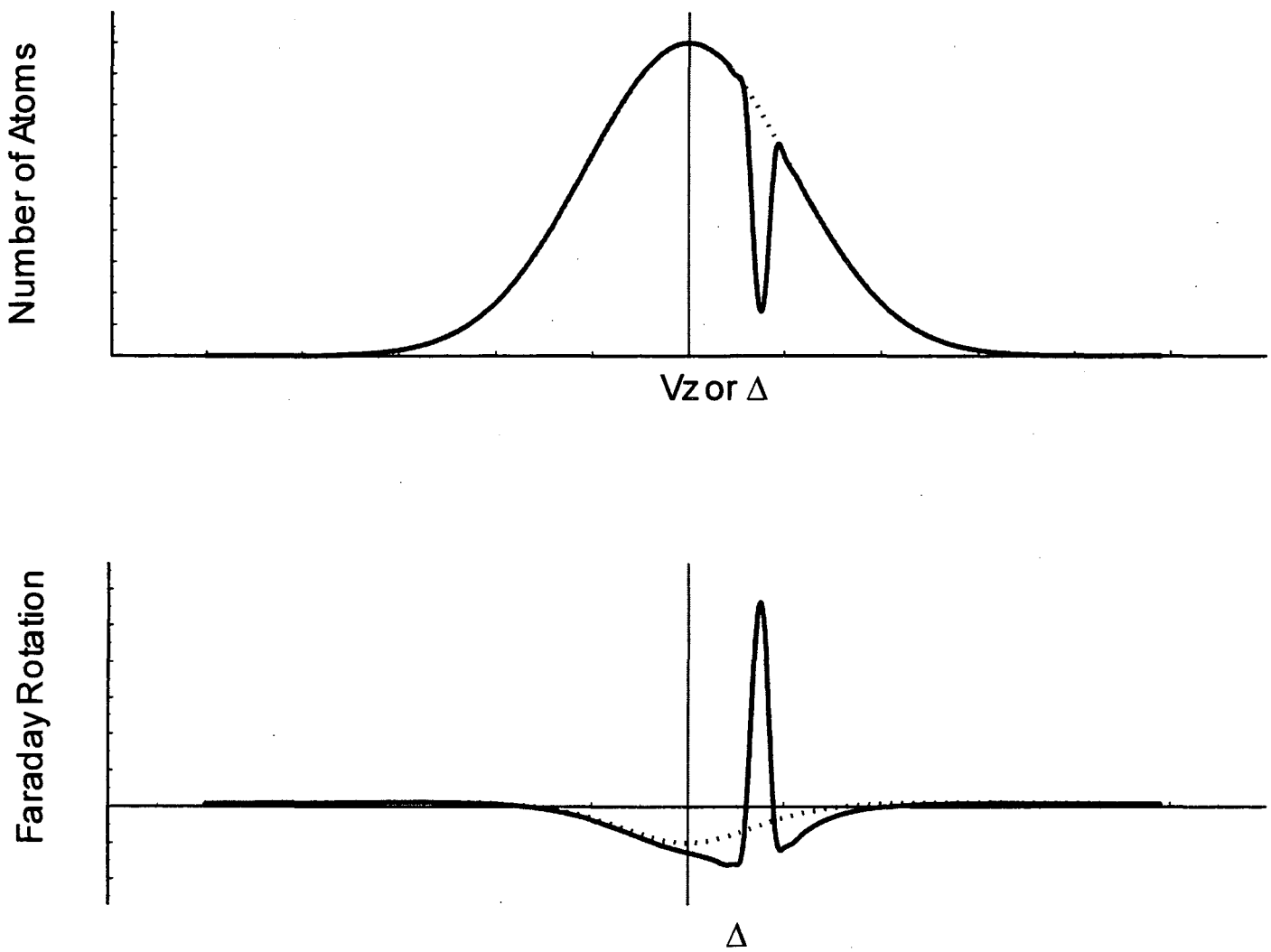


Fig. 4. The hole-burning effect in nonlinear Faraday rotation. This figure assumes $|g\mu_B| \ll \Gamma$. Monochromatic laser light produces a Bennett hole in the velocity distribution of atoms in the lower state of the optical transition (upper trace). The Faraday rotation produced by atoms with such velocity distribution can be seen as rotation produced by unperturbed distribution minus the rotation that would have been produced by atoms removed by optical pumping (lower trace).

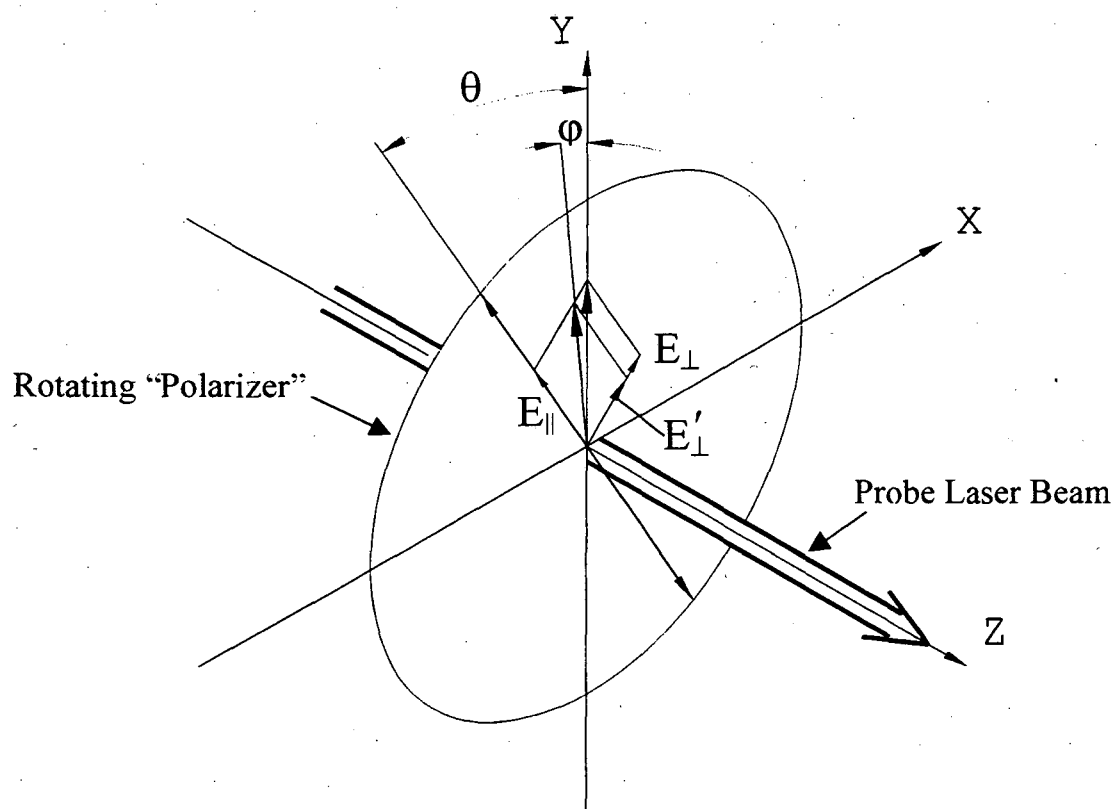


Fig. 5. An optically thin sample of aligned atoms precessing in a magnetic field can be thought of as a thin rotating Polaroid film, which is transparent to light polarized along its axis (E_{\parallel}), and slightly absorbent for the orthogonal polarization (E_{\perp}). The effect of such "polarizer" is to rotate light polarization by an angle $\phi \propto \sin(2\theta)$.

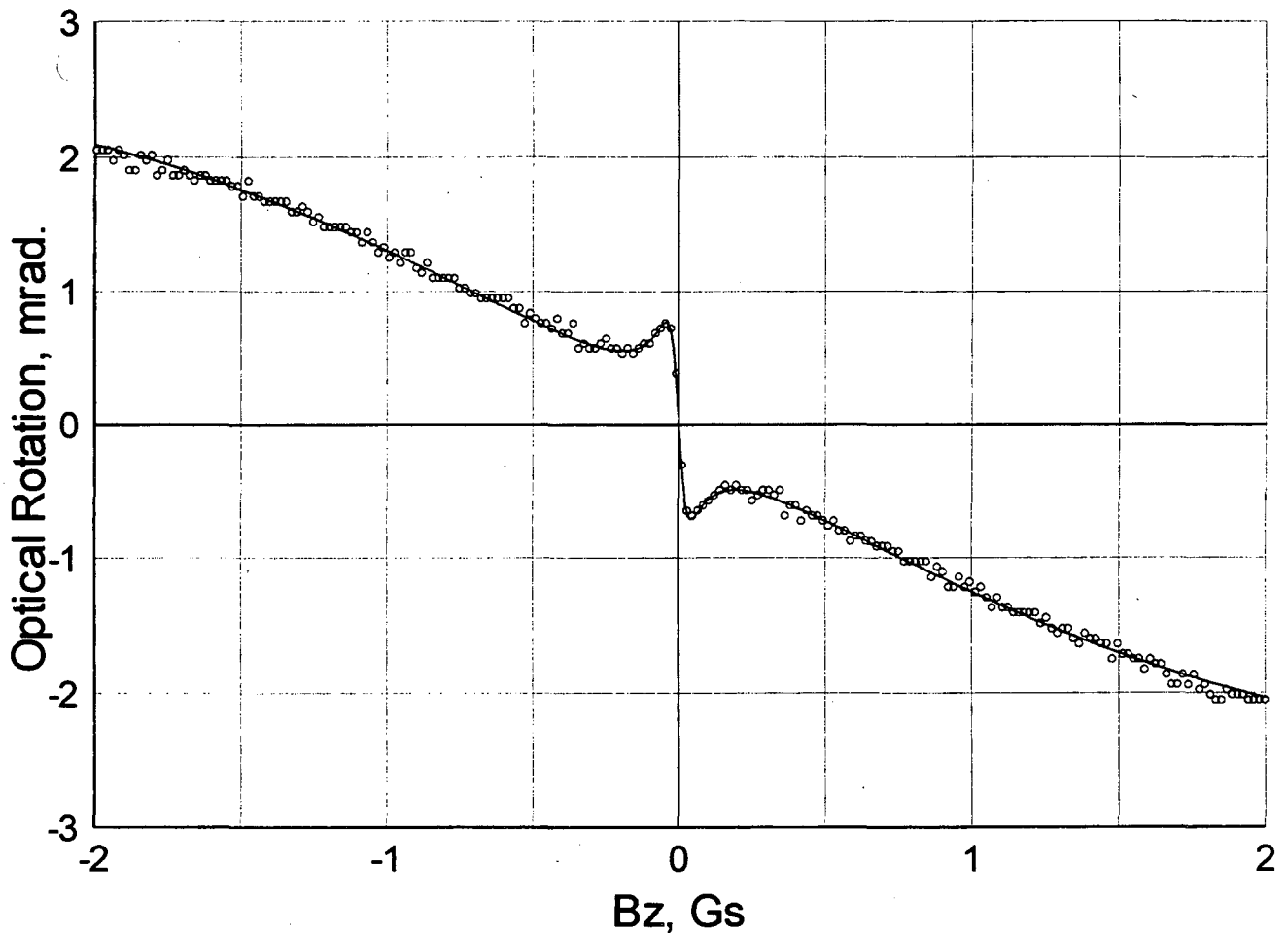


Fig. 6. Optical rotation dependence on the longitudinal magnetic field. The broader (clipped) resonance corresponds to the hole-burning effect, while the narrower resonance is due to the coherence effect with effective relaxation due to atoms' transit across a laser beam (effective diameter ~ 3 mm). Laser power: $15 \mu\text{W}$ (the optical pumping saturation power is $\approx 150 \mu\text{W}$; the magnitude of the rotation increases with power approximately linearly up to the saturation power). An additional resonance can be seen in a detailed scan of the near-zero magnetic field region (see Fig. 7). The solid line is a fit to a theoretical lineshape described in the text.

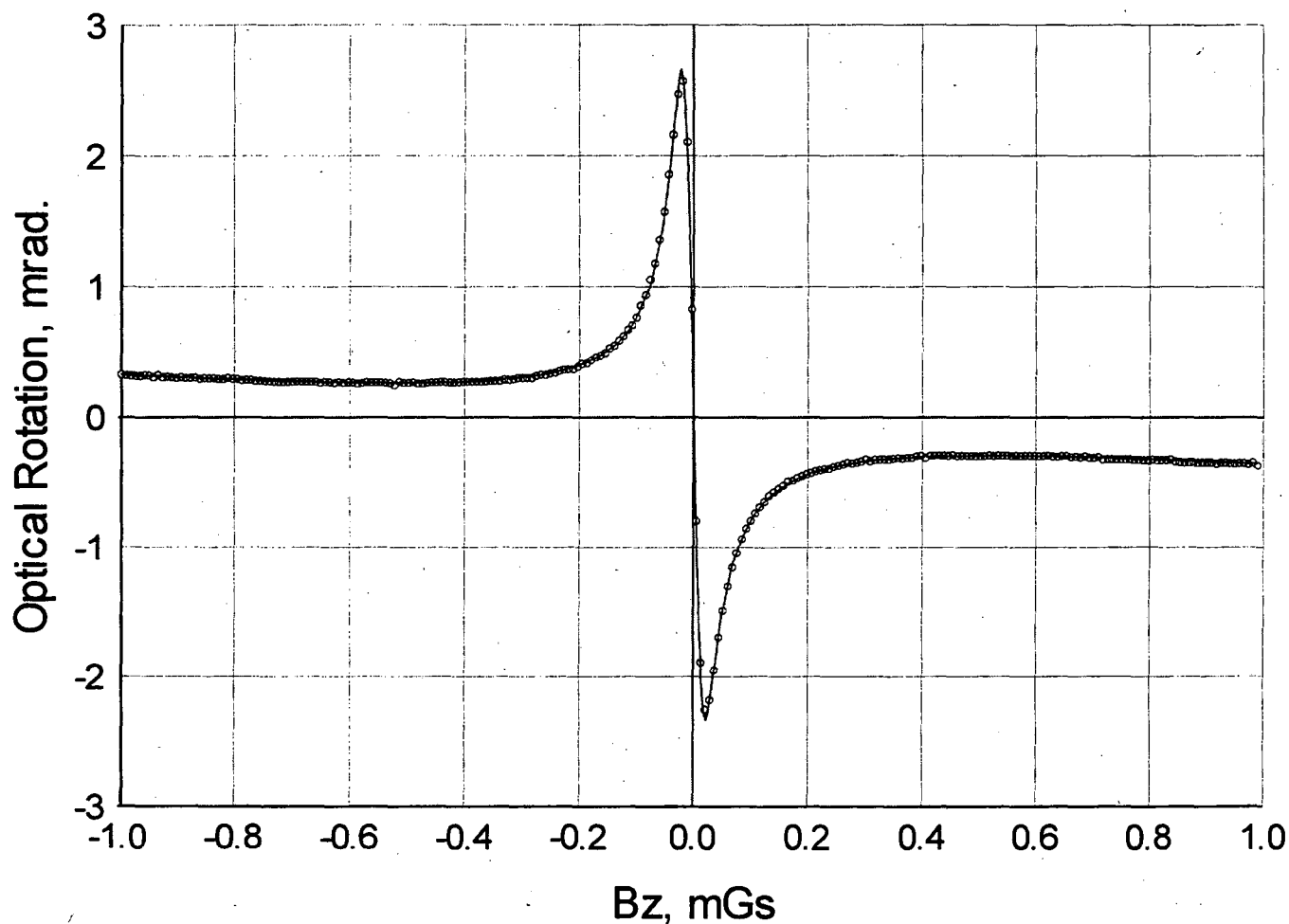


Fig.7. A detailed scan of the near-zero magnetic field region showing the resonance due to the coherence effect with relaxation time corresponding to multiple wall collisions. To our knowledge, this is the narrowest resonance in magnetic field dependence of the optical rotation ($40 \mu\text{Gs}$ peak-to-peak) observed so far. The steep derivative of the rotation near the center provides for a high sensitivity to small magnetic fields. Laser power: $100 \mu\text{W}$. The slight asymmetry of the resonance is due to a residual transverse magnetic field of a few μGs .

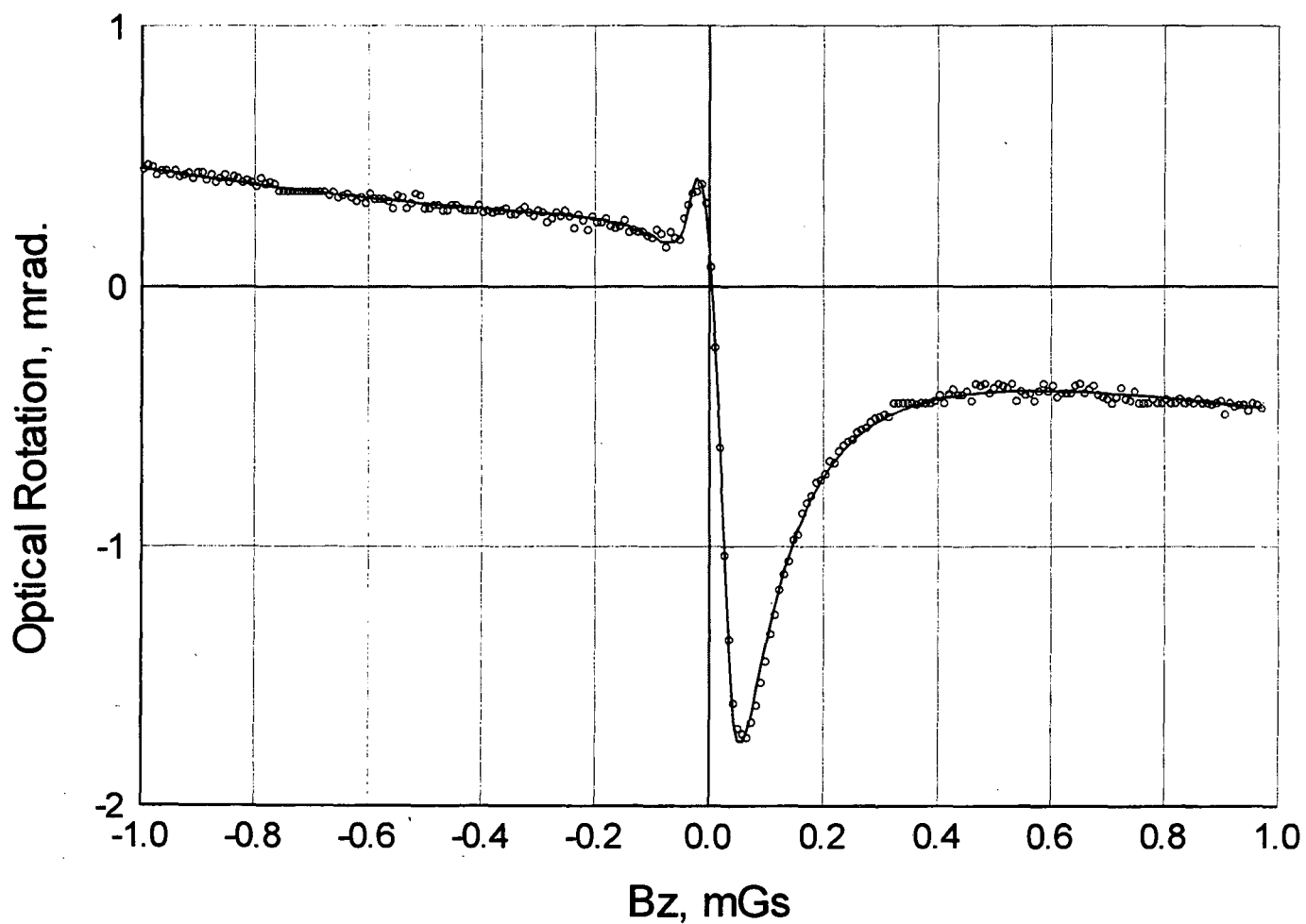


Fig. 8. An example of the longitudinal magnetic field scan in the presence of applied transverse fields: $B_x=43.5 \mu\text{Gs}$, $B_y=65.2 \mu\text{Gs}$. Laser power: $89 \mu\text{W}$. Solid line is a fit with a simple theoretical model described in the text. Since the lineshape is sensitive to all components of the magnetic field, this method can be applied to sensitive 3-axis magnetometry.

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