Level-crossing experiments by two-photon excitation

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Abstract. Complete orientation of the Tl 7 2P3/2(F = 2) state by two-photon excitation allows Hanle-effect measurement in direct fluorescence as well as in the optical radiation cascade. The experiment provides estimations either of both the lifetimes of the involved excited states, or, if one of the lifetimes is reliably known, two independent determinations of the other. The measurements are reported on the background of other nonlinear Hanle-effect and level-crossing experiments.

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1. Introduction

Since the very early Hanle-effect measurement [1] of a Doppler-free level-crossing signal detected in polarized lateral fluorescence of optically excited and polarized Hg 3P1 states, level-crossing experiments have been carried out in various atomic, particle, or solid systems and under very different conditions of anisotropic excitation and detection in external or internal, static or dynamic, magnetic or electric fields [2, 3].

Lasers [4] with narrow lines and high radiation intensities allow the generation of polarizations by linear and nonlinear light interaction and, furthermore, detection of Hanle-signals in absorption as well as in stimulated emission. The Hanle-effect of polarized atomic levels populated by stimulated emission can be measured in lateral fluorescence; nonlinear (or stimulated) Hanle-effect and high-field level-crossings can be probed by laser light via polarization spectroscopy and mode-crossing experiments. Thus, by nonlinear laser techniques and by developing analytical applications [5] during the 70th and 80th Prof. Hanle’s former institute still won interesting results by new methods of optically excited level- and line-crossing [6] based on the thesis of its senior director.

In a previous paper [7] Hanle-effect and level-crossing measurements at 8 2P3/2- and 9 2P3/2-levels of thallium obtained by pulsed two-photon excitation were reported. The effect owing to alignment can only be found in direct fluorescence of the polarized levels. The two-photon excitation however, allows complete orientation of the n 2P3/2(F = 2)-states of thallium. Hunter et al. [8] measured Hanle-effect in the cascade by pulsed two-photon excited orientation of the 7 2P3/2(F = 2) state on the transition 7 2S1/2(F = 1) → 6 2P1/2(F = 0). The signal resulting from this measurement is a product of two Hanle-curves with the width combining both lifetimes. In this paper, Hanle-effect measurements are reported with excitation by cw-laser two-photon absorption and observation both in the emission via the direct transition 7 2P3/2(F = 2) → 7 2S1/2(F = 1) (on λ = 1151 nm), and in the cascade via the 7 2S1/2(F = 1) → 6 2P1/2(F = 0) transition (on λ = 377.5 nm).

Since various linear and nonlinear Hanle-effect phenomena are known, which result from two-photon interaction in absorption as well as in stimulated emission, a clarifying basic discussion is given with experimental examples.

2. Basics of Hanle-effect in two-photon interaction

2.1. Polarized saturation of levels by Raman-like two-photon interaction

Atomic states can be populated and polarized by absorption as well as by stimulated transitions from a higher level. And, polarization of a state (a) by an optical transition can be also detected in absorption or in stimulated emission if the polarization contributes relevantly to the saturation of the transition, i.e. if the polarization, ρa(1) or ρa(2) (orientation or alignment), is not negligible compared with the population difference ρa(0) − ρa(0). Since all atoms interacting with the radiation field of a narrow laser line belong to the same homogeneous velocity ensemble, the saturated Hanle-signal is Doppler-free, but
due to equal path lengths in forward direction [9] owing to a macroscopic coherence of the atomic ensemble. However, under conditions of a linear approximation this macroscopically coherent interaction results in Doppler-broadened level- and line-crossing [6] phenomena.

Nonlinear Hanle-effect detected by polarization spectroscopy. A "nonlinear", "saturated" or "stimulated Hanle-effect" has been measured by a very early experiment [10] of nonlinear polarization spectroscopy [4, 11]. When an axial magnetic field is applied to a gas laser with Brewster-angled windows, the plane of polarization is rotated by the Faraday-effect with the angle of rotation following the Faraday function [12], i.e. a Doppler-broadened dispersion profile. However, owing to the saturation, which results from changes of population and of atomic polarization, additional narrow signals are found at zero- and non-zero field strengths (Fig. 1) [13]. The first is the nonlinear Hanle-effect, the latter can be interpreted as the related dynamic effect which occurs, when, at mode-crossing magnetic field strengths (see next paragraph), the Larmor precession frequency of the atomic polarization is equal to the laser-radiation beat-frequency. Then the rotating anisotropy enhances the saturation effect of the population. The width $\Delta B$ of these nonlinear signals is given by the relaxation constant $\gamma_a(2)$ of the atomic polarization $(g_a(c/2m) \Delta B = 2\gamma_a(2))$.

Detection via laser intensity. At magnetic fields of atomic level-crossings ($\delta B = 1$ or 2, depending on the direction of magnetic field), or at field strengths, where the splitting of those atomic levels equals the separation of different laser modes [14, 15] ("mode-crossing" [16]), the same atoms are simultaneously saturated by two components of counterpolarized light. Saturation effects of population as well as of polarization reduce atomic absorption or amplification of light. Thus, nonlinear Hanle-effect and level-crossing, zero- and high-order mode-crossing can be detected by the effect on light polarization as well as on light gain or loss in an atomic system. Figure 2 shows mode-crossing signals measured by laser intensity in high order and, thus, allowing high resolution Zeeman spectroscopy [17]. Hyperfine structure high-field level-crossings can also be measured by zero-order mode-crossing experiments (Fig. 3) [18].

The atomic polarization is created and detected by the interaction of two photons connecting two Zeeman-sublevels of one of the states (a) by optical-optical double resonance [2, 19] via an intermediate state (b) (upper Fig. 2) [13]. One photon is creating, the other is probing the saturation. In absence of atomic collisions, thereby, the Zeeman-tuned width of this Raman-like [20] folded two-photon transition only depends on the relaxation constant $\gamma_a$ of the coherence of the initial and final Zee-