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► To cite this version:

D. Lecler, R. Ostermann, W. Lange, J. Luther. Dependence of Hanle signals on the spectral distribution of the exciting light. Journal de Physique, 1975, 36 (7-8), pp.647-651. 10.1051/jphys:01975003607-8064700 . jpa-00208298

HAL Id: jpa-00208298

<https://hal.science/jpa-00208298>

Submitted on 1 Jan 1975

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Classification
 Physics Abstracts
 5.250

DEPENDENCE OF HANLE SIGNALS ON THE SPECTRAL DISTRIBUTION OF THE EXCITING LIGHT

D. LECLER (*), R. OSTERMANN (**), W. LANGE (***) and J. LUTHER (****)

Institut A für Experimentalphysik der T.U. Hannover, Germany

(Reçu le 27 février 1975, accepté le 18 mars 1975)

Résumé. — On peut observer des courbes d'effet Hanle très déformées si la largeur naturelle du niveau étudié n'est pas très faible devant la largeur Doppler et si le profil spectral de la source excitatrice n'est pas plat à l'échelle de la largeur naturelle. Une expérience Hanle de ce genre a été réalisée sur le niveau de résonance du calcium ; la raie excitatrice émise par une lampe à cathode creuse est filtrée par un jet atomique de calcium qui creuse dans le profil spectral un *trou* très étroit à la fréquence de résonance. Les effets observés sont très bien expliqués par la théorie.

Abstract. — The shape of Hanle (zero field level crossing) signals may be strongly distorted if the spectral profile of the exciting light varies rapidly on the scale of the natural linewidth, provided that the Doppler width in the scattering region is not very large compared with the natural width. A Hanle experiment has been performed at the resonance line of calcium. The exciting light had a spectral profile with a very narrow *hole* at the resonance frequency and was produced by a calcium hollow-cathode with an absorbing atomic beam of calcium inserted in its light path. The observed level crossing signals are in very good agreement with theoretical predictions.

1. Introduction. — The frequency distribution of atomic fluorescent radiation in the case of non broad-band excitation has been widely discussed and investigated [1, 2]. If the atoms are coherently excited, the polarization of the emitted light will depend on the frequency distribution of the exciting light source. Under these conditions the shape of Hanle signals must deviate from the well known Lorentzian shape (or dispersion shape) that one obtains when using broad line excitation and detection [3, 4].

The recent development of narrow band dye lasers gives new interest to this problem. Atomic fluorescence and level crossing with monochromatic excitation are now well studied [e.g. 5, 6]. Hanle experiments using narrow band lasers have also been recently reported [7, 8].

The purpose of this note is to point out that deviations of Hanle signals from their usual shape can occur not only with lasers but also with conventional light sources [see e.g. 9], for example due to strong self reversal of the exciting light source.

In order to study these influences we have performed experiments in which the ($4s\ 4p\ ^1P_1$) level of ^{40}Ca (resonance transition) was excited by light with a broad intensity distribution that has a well defined sharp hole at the resonance frequency, produced by a *resonant filter*. The shape of Hanle curves has been studied experimentally and compared with theoretically calculated signals.

2. Experimental procedure. — In order to fulfil the required conditions for observing perturbed Hanle curves, the following special experimental conditions were chosen :

2.1 A SHORT LIVED EXCITED STATE. — The experiments were performed on the calcium resonance line ($\lambda = 4\ 227\ \text{\AA}$) $4\ s^2\ ^1S_0 - 4s\ 4p\ ^1P_1$. The lifetime of the 1P_1 state is rather short ($\sim 5\ \text{ns}$ [10]) so the natural line width of the transition is large ($\sim 1.2\ \text{mK}$). Since natural calcium contains 97 % of the even isotope ^{40}Ca there are no complications due to hyperfine splittings or isotope shifts.

2.2 A SMALL DOPPLER WIDTH. — In our experiments the light was scattered by a collimated beam of Ca atoms (Fig. 1). The Doppler width in the directions of excitation and observation was about 7 mK (collimation ratio of 1/20). This value represents a compromise between the requirements of a small Doppler broadening and of a sufficient density of atoms in the scattering region.

(*) Laboratoire de Spectroscopie Atomique (LA 19), 14032 Caen Cedex, France.

(**) Universität Marburg, 355 Marburg/Lahn Renthof, 5 Germany.

(***) Fakultät für Physik der Universität Bielefeld, 48 Bielefeld Viktoriastr., 44 Germany.

(****) Fachbereich IV Universität Oldenburg, 29 Oldenburg Ammerländer Heerstr., 98 Germany.

FIG. 1. — Schematic diagram of the experimental set up.

2.3 A SHARP RESONANT FILTER IN THE LIGHT PATH. — A highly collimated atomic beam absorber was inserted in the light path as an extremely sharp resonant filter (Fig. 1). In order to get enough absorption we used an arrangement of 30 parallel atomic beams with collimation ratio of 1/50 in the direction of the incident light. Typical data of the filter are : length of the absorbing region 10 cm, density of atoms 10^9 cm^{-3} .

The experimental set up, schematically represented in figure 1, is a typical level crossing arrangement. The only peculiarities are the atomic beam absorber and a Fabry-Perot interferometer, used to monitor the profile of the light source (without absorber). Geometrical conditions for excitation and detection are summarized in figure 2 the magnetic field H and the direction of excitation are in the Oz direction. The light is detected in the Ox direction. A rotating polarizer at an angle $\alpha(t)$ with respect to Ox is used in the excitation light path.

FIG. 2. — Geometrical conditions for excitation and detection.

3. Results. — It has been shown from general considerations on polarized light [11] that the intensity S_T of the fluorescence in the Ox direction can be written.

$$S_T(H) = S_I(H) - S_A(H) \cos 2\alpha - S_D(H) \sin 2\alpha \quad (1)$$

$S_I(H)$ is the incoherent part of the signal ; $S_A(H)$ is the *absorption shaped* signal ; $S_D(H)$ is the *dispersion shaped* signal.

For broad line excitation and detection. S_I is a constant, $S_A = 1/(1 + X^2)$ and $S_D = X/(1 + X^2)$ where $X = H/H_{1/2}$ and $H_{1/2} = \Gamma/2 \gamma = 1/2 g \mu_B \mathcal{G}$ ($\mathcal{G} = 1/\Gamma$ is the lifetime ; g the lande factor and μ_B the Bohr magneton).

These three signals can be easily monitored with a rotating polarizer and lock-in techniques [11], S_I is proportional to the D.C. signal and the signals S_A and S_D are separated by proper settings of the reference phase of the lock-in amplifier. Figures 3, 4, 5 give examples of the results obtained for S_I curve *c*, S_A curve *a* and S_D curve *b* respectively using various atomic densities in the absorber. The unit for the H axis is the half width $H_{1/2}$ determined in a Hanle experiment without absorber. The atomic density in the scattering beam has been chosen low enough to avoid multiple scattering. Figure 3 corresponds to a weak absorption and figures 4 and 5 to increased atomic densities in the absorber. The points which are calculated using the theory summarized in the next paragraph are in quite good agreement with the experiments. The curve *a* on figure 3 crosses the horizontal axis. The reasons are the relatively small line width of the hollow cathode source cooled with liquid nitrogen and a small polarization (due to reflection on mirrors) of the light even before passing through the rotating polarizer. These conditions have been taken into account in the theoretical calculations.

It will be shown theoretically in the next section that the discrepancy between *anormal* and *normal* Hanle curves is not only due to magnetic scanning (represented by the S_I signal) but also to dispersion effects of the scattering atoms (see Fig. 6).

4. Theory. — The intensity of the light scattered by a motionless atom with angular momentum $J = 0$ in the ground state and $J = 1$ in the resonance state, excited by a monochromatic radiation of angular frequency k_i can be calculated by applying to this particular case the general theory of resonance fluorescence developed by Heitler [1]. Cohen-Tannoudji [6] has shown that the same results are also obtained in the limiting case of low intensity excitation of atoms by monochromatic laser light. We use the same methods to calculate the intensity of the scattered light at right angles from the excitation in the following way :

FIG. 3.

FIG. 4.

FIG. 5.

FIG. 3, 4, 5. — Examples of perturbed Hanle signals for various atomic densities in the absorber. The effective Doppler widths in natural width units are : in the absorber $\delta\omega_D/\Gamma = 3$ and in the scattering beam $\Delta\omega_D/\Gamma = 6$. On each figure curve *a* represents the signal $S_A(X)/S_A(0)$; curve *b* : $S_D(X)/S_A(0)$; curve *c* : $S_I(X)/S_I(0)$ and curve *d* : the plot of $(S_I - S_A)/S_D$. All these curves have been represented for $H/H_{1/2} = X > 0$.

FIG. 6. — Plot of calculated functions S_A/S_1 normalized to 1 for $H = 0$. In full line, perturbed Hanle signal, in dotted line normal Lorentzian shape Hanle curve.

4.1 We assume that the excitation is a sum of monochromatic waves of angular frequency k_i completely incoherent with each other; we call $I(k_i)$ the frequency distribution of the light intensity.

4.2 We suppose that the detection is of the *broad band* type.

4.3 We calculate the intensity $i(\omega)$ scattered by the atoms having a component of velocity v_i in the direction of excitation when a given magnetic field H is applied ($\omega = \gamma H$ is the Larmor frequency).

4.4 We compute the sum of the intensities scattered by atoms having different velocity components on the direction of excitation. In an atomic beam (e.g. see [12], p. 90) the angular frequency distribution $F(a_i) da_i$ of the Doppler shifts $a_i = k_i v_i/c$ in a direction perpendicular to the beam can be approximately represented by

$$F(a_i) da_i = \frac{1}{u_i \sqrt{\pi}} \exp(-a_i^2/u_i^2) da_i$$

$$u_i = \frac{\widetilde{\Delta\omega_D}}{2\sqrt{\ln 2}} = \frac{s \Delta\omega_D}{2\sqrt{\ln 2}};$$

$\widetilde{\Delta\omega_D}$ is the effective Doppler width, s is the collimation ratio of the beam.

All these steps are very similar to those used to interpret the effect of Faraday rotation on the shape of Hanle signals [13]. The result can be conveniently written in the following form :

$$I(\omega) = I_0 \sum_{m,m'} A_{m,m'} \frac{\Gamma}{(m - m')\omega - i\Gamma} \times$$

$$\times \left\{ (P_m - P_{m'}) - \frac{i}{2} (Q_m + Q_{m'}) \right\}. \quad (2)$$

In this equation m, m' are the Zeeman quantum

numbers ($m = 0, \pm 1$) of the excited state; $A_{m,m'}$ is a product of dipole matrix elements under the geometrical conditions of excitation and detection [3, 4, 13] (*); Γ is the inverse of the coherence time of the excited level (including possible effects of multiple light scattering or collisions); the functions P_m and Q_m are defined by the equation :

$$P_m + iQ_m = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} da_i dk_i I(k_i) \times$$

$$\times F(a_i)/(k_i - a_i - k_0 - m\omega + i\Gamma/2) \quad (3)$$

(k_0 is the resonance angular frequency).

These functions must be evaluated in most cases by numerical integration of eq. (3). It is helpful to perform just the integration over a_i using the plasma dispersion function [13, 14].

Under the geometrical conditions of our experiments we use eq. (2) to compute the signals defined in eq. (1) and we obtain :

$$S_I(X) = (Q_- + Q_+)/2 Q_0 \quad (4a)$$

$$S_A(X) = [X(P_- - P_+) + (Q_- + Q_+)/2 Q_0(X^2 + 1)] \quad (4b)$$

$$S_D(X) = [X(Q_- + Q_+) - (P_- - P_+)/2 Q_0(X^2 + 1)] \quad (4c)$$

we have written

$$P_{m=0,\pm 1} = P_{0,\pm}, \quad Q_{m=0,\pm 1} = Q_{0,\pm},$$

$$X = H/H_{1/2} = 2 \gamma H \Gamma = 2 \gamma H/\Gamma.$$

With the aid of a computer program we calculate :

1) The intensity distribution $I(k_i)$ of the exciting light. We assume that the line profile $I_0(k_i)$ of the hollow cathode is gaussian with a half width measured by Fabry-Perot interferometry. The absorber *burns* a hole in this profile and the new one $I(k_i)$ is calculated, knowing the atomic density N and the effective Doppler width $\widetilde{\delta\omega_D}$ in the absorption beam.

2) The integrals appearing in eq. (3).

3) The signals given by eq. (4).

The important parameters of the experiment : N_a (atomic density in absorber), $\widetilde{\delta\omega_D}/\Gamma$ (ratio of the effective Doppler width in the absorber to the natural width), $\widetilde{\Delta\omega_D}/\Gamma$ (ratio of the effective Doppler width in the scattering beam to the natural width) have been adjusted to get the best fit of the theoretical points with the experimental curves. The values of these parameters agree quite well with the experimental ones calculated from the temperature of the atomic beam ovens and the collimation ratios. In figures 3, 4, 5 only the atomic density in the absorber beam has been

(*) In the denominator of the last factor of eq. (1) of reference [13] one should read $+ a_i$ instead of $- a$.

changed (Fig. 3 : $N_a = 2.5 \times 10^8 \text{ cm}^{-3}$; Fig. 4 : $N_a = 1.6 \times 10^9 \text{ cm}^{-3}$; Fig. 5 : $N_a = 3 \times 10^9 \text{ cm}^{-3}$).

The good agreement between the experimental and the calculated curves proves the validity of the hypotheses made in our calculation. A remarkable property of eq. (4) is the relation

$$[S_I(X) - S_A(X)]/S_D(X) = X = H/H_{1/2}.$$

We have plotted this ratio on curve *d* of figures 3, 4, 5. We verify that with the choice of the *H* scale we get a straight line of slope unity. This relation holds independently of $I(k_i)$, which makes it possible to determine $H_{1/2}$ with any intensity distribution provided that S_I , S_A and S_D can be measured as functions of *H*.

The deformation of Hanle signals is not only due to the magnetic scanning represented by the incoherent signal S_I . This signal depends only on the absorption properties of the vapour represented by the real functions Q . On the contrary the coherent signals S_A and S_D depend also on the dispersion properties of the scattering vapour represented by the *P* functions. It can be seen in figure 6 that the ratio S_A/S_I (full line) differs from the lorentzian curve (dotted line) that would have been obtained if the deformation of Hanle signals were due only to magnetic scanning effects.

5. Conclusion. — The magnetic depolarization curves can be interpreted in the frame of Heitler's

theory in our case (sharp hole burned in a broad line) as well as in the case of low intensity monochromatic excitation by laser light. We have established the eq. (2) and (3) which allow to calculate the signals under any geometrical conditions with any intensity distribution of the exciting light source. One finds easily from (2) and (3), which are the conditions to get perturbed Hanle curves and possible systematic errors in the determination of coherence times from the width of such curves, that such difficulties occur if the functions P_m and Q_m (3) vary noticeably with ω (i.e. with *H*) in a range where the light is not completely depolarized (ω not too large with respect to Γ). From a physical point of view two conditions are required :

1) The intensity distribution $I(k_i)$ has strong variations in the region of the resonance frequency $k_i \simeq k_0$.

2) The Doppler width of the scattering atoms is the same order of magnitude as the natural width of the transition involved ; this point has been stressed by Series [15]. When both these conditions are fulfilled or nearly fulfilled, experimentalists should be rather cautious in extracting coherence times from the shape of observed Hanle curves.

Acknowledgment. — We are very grateful to Professor A. Steudel, Director of the « Institut A Für Experimentalphysik der T.U Hannover », who enabled us to do these experiments in his institute.

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