COHERENCE NARROWING AND SHIFT OF OPTICAL DOUBLE RESONANCE SIGNALS IN THE (4 s 4p) 1P1 STATE OF Ca AT HIGH MAGNETIC FIELD

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COHERENCE NARROWING AND SHIFT OF OPTICAL DOUBLE RESONANCE SIGNALS IN THE (4*s* 4*p*) $^1P_1$ STATE OF Ca AT HIGH MAGNETIC FIELD

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Résumé. — Nous avons observé l'influence de la diffusion multiple de la raie de résonance sur les signaux de double résonance optique dans un jet atomique de Ca pour une fréquence de Larmor de 2,5 GHz. L'expérience a donné une largeur minimum $\Delta\nu_{\lim} = \left(0{,}26 \pm {}^{0{,}0}_{0{,}06}\right) \Delta\nu_{nat}$ et un déplacement de fréquence dû au temps de vol du photon égal au maximum à $\delta\nu/\nu = -0{,}28\,\%$.

Abstract. — The influence of multiple scattering of resonance radiation on ODR signals was observed in a dense atomic beam of Ca at a Zeeman splitting of 2.5 GHz. The experiment yielded a limit of narrowing $\Delta\nu_{\lim} = \left(0.26 \pm {}^{0.0}_{0.06}\right) \Delta\nu_{nat}$ and a maximum time of flight shift $\delta\nu/\nu = -0.28\,\%$.

Some phenomena of multiple coherent scattering [1] can be observed preferably by ODR at high magnetic fields, namely [2] :

(1) The suppression of T_1 relaxations during coherence transfer by satisfying the condition that the Zeeman splitting is large compared to the Dopplerwidth. (2) The retardation of the average Larmor precession by the time of flight of the photon. (3) The acceleration of the average Larmor precession by the Faraday effect. (4) The indication of relaxation due to the stochastic character of (2) and (3).

In previous papers [3] [5] experiments on this subjects were reported which were conducted on the $^3P_1$ state of ^{198}Hg. The results are briefly summarized here : (1′) Minimum observed linewidth $\Delta\nu_{min} = 0.23\ \Delta\nu_{nat}$ (theoretical limit : $\Delta\nu_{min} = 0.12\ \Delta\nu_{nat}$). (2′) Maximum observed time of flight shift $\delta\nu/\nu = -4.5 \times 10^{-5}$. (3′) Maximum observed Faraday shift

$$\Delta\nu/\nu = +4.5 \times 10^{-5}\,.$$

(4′) At the highest frequencies and medium densities the deviation of (1′) from theory could be explained by time of flight relaxation. (2′) and (3′) are in satisfying agreement with theory.

For the following reasons it was desired to repeat these experiments in a short lived state which is connected to the ground state by a strong resonance line : a) Since such an experiment would run at lower densities, pressure broadening which could not be excluded in the case of Hg would be avoided safely. b) The relative frequency shifts were expected to increase as the ratio $\Gamma(^1P_1$, Ca) over $\Gamma(^3P_1$, Hg) of the spontaneous transition probabilities.

The extension of this kind of experiment to a short lived state implies severe experimental complications, however. The energy density of microwaves in the cavity has to be increased proportional to the square of the natural linewidth in order to achieve a constant transition probability. First attemps on the $^1P_1$ state of Hg ($\Delta\nu = 240$ MHz) and Cd ($\Delta\nu = 160$ MHz) were unsuccessful because of this problem. Therefore the $^1P_1$ state of Ca ($\Delta\nu = 71$ MHz) was finally chosen which is excited from $4\,s^2\ ^1S_0$ groundstate by the resonance line ($\lambda = 4\,227$ Å, $f = 1.8$).

The experimental set-up is shown in figure 1. An atomic beam of Ca which crossed a microwave cavity served as absorbing medium. This solution of the resonance vessel problem, however, did not permit a reliable calibration of the density. Moreover the latter does not obey a Boltzmann distribution, which is usually considered in the theory (*). Microwave power of max 100 W at 2.5 GHz was supplied by a magnetron Valvo 7090 to the cavity. The frequency stability of this system — about 2 % under usual conditions — was increased to 1×10^{-5}/min by careful stabilization of the dc power supply and especially by strong coupling to the cavity, since the bandwidth of the full system is drastically reduced by this

(*) By use of a MgO resonance cell [6] these problems would have been avoided. But it seemed doubtful that a sufficient microwawe field could have been achieved in this complicated, hot system.

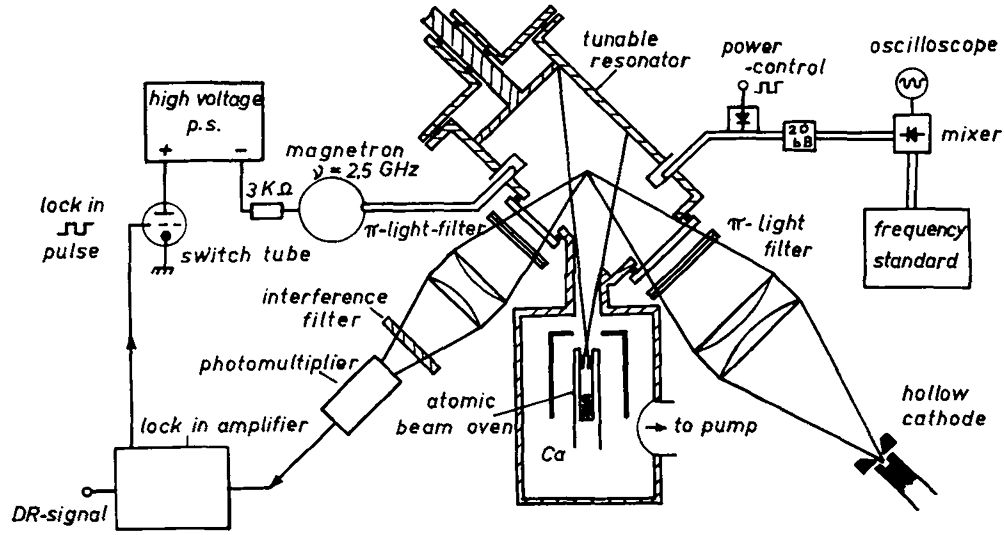

FIG. 1. — Diagram of the apparatus

step. The incident and scattered light beam were directed at angles of 45° and 135° resp. to the atomic beam, all three of them in the plane of the pole gap. The diameter of the beam at the place of observation was about 4 cm. A hollow cathode served as a light source. Because of the Dopplershift the center of π excitation of the beam just coincided with one of the bumps of the slightly self-reversed spectral line. ODR-signals were observed by lock in amplification of the change of the scattered π light intensity, which was caused by a square-wave modulation of the magnetron voltage.

The signals were recorded by sweeping the magnetic field through the resonance. The field was calibrated by proton resonance at either side of each ODR-curve. Useful signal to noise ratios were achieved with time constants of 1 s. Width and center of the ODR curves were determined numerically by a least square fit to a Lorentzian. Figure (2) shows the results in a plot of the resonance shift ($g_{J\,\mathrm{eff}}$) versus the resonance narrowing ($\Delta\nu_{\mathrm{eff}}/\Delta\nu_{\mathrm{nat}}$). With only a few points the power broadening could be checked by extrapolating to zero power. At very high densities, however, the experimental conditions could not be kept constant for a time long enough for an application of this technique. At the present time, therefore, an uncertainty in the power broadening of the linewidths up to max 15 % cannot be excluded at the high pressure side of the plot.

The plot in figure 2 is especially suitable for the interpretation of this experiment, since it does not require the knowledge of the optical thickness of

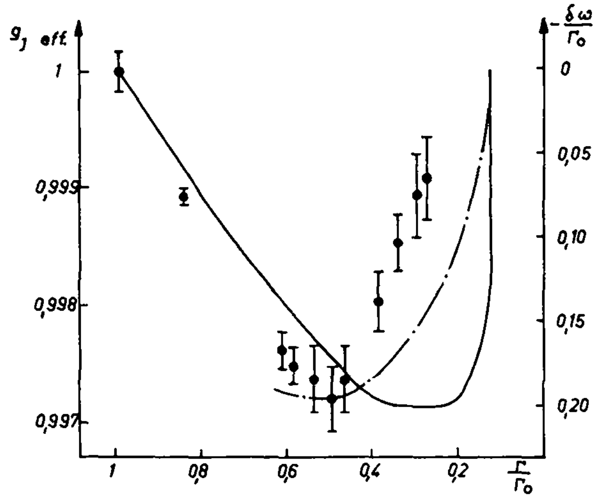

FIG. 2. — Line shift versus line narrowing of the DR signal at 2.5 GHz in ^{40}Ca ($4s4p\ ^1P_1 \rightarrow 4s^2\ ^1S_1$) and line shift versus line narrowing calculated from theory :

——— not including time of flight relaxation.

— · — including time of flight relaxation.

Density rises from left to right.

the beam. However, the following qualitative observations regarding this quantity are important : (1) On the right side of the plot the narrowing reaches an asymptotic value as a function of the temperature of the atomic beam oven. (2) From the temperature of the oven the density can be roughly estimated to reach 10^{12} cm^{-3} in this region, a value at which theoretically the narrowing should be saturated. Therefore it can be stated, that in this experiment an asymptotic linewidth of $\Delta\nu_{mn} = (0.26 \pm {}^{0}_{0.06}) \times \Delta\nu_{nat}$ is observed. It is remarkable that this number agrees within the limit of error with the one obtained in mercury under completely different conditions. The time of flight shift reaches a maximum of -0.28% which is a factor of 60 larger than in the Hg-experiment. Under identical geometrical conditions one expects theoretically this ratio to be equal

$$\Delta\nu_{nat}(\mathrm{Ca})/\Delta\nu_{nat}(\mathrm{Hg}) = 26 .$$

The difference is due to the longer total diffusion length in the Ca-experiment. No Faraday effect is observed either as a consequence of insufficient vapor pressure or of the spectral shape or of both.

The solid curve in figure 2 is taken from a calculation [4] of the narrowing and time of flight shift as functions of the optical thickness (NKR) following the theory outlined by Omont [2]. The maximum shift is fitted. It clearly shows that beyond a certain density of the absorbing medium the narrowing is not as strong as expected due to some relaxation phenomenon not put into account by the theory. At medium densities where the time-of-flight-effect is important, the discrepancy may be partly attributed to a relaxation related to this effect : Since the time of flight between emission and reabsorption of a photon in the vapor shows a statistical distribution, the time-of-flight-effect is accompanied by a destruction of coherence in multiple scattering. A calculation in the frame of the theory [4], [7] that puts into account this relaxation leads to the dotted curve. It is not extended to lower densities since the approximation $\omega . T \ll \pi$ ($T =$ time of flight), made in the calculation is no longer valid there. As for the higher densities it should be pointed out that the current theories consider relaxation in the process of multiple scattering only by the absorption of light neglecting the effect of dispersion. The observation of twice the theoretical relaxation rate leads to the suggestion, therefore, that the dispersion contributes to the relaxation as much as the absorption.

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References

[1] BARRAT (J. P.), *J. Physique Rad.*, 1959, **20**, 541, 633, 657.
[2] OMONT (A.), *J. Physique*, 1965, **26**, 576.
Thèse de Doctorat d'Etat, Faculté de Sciences de Paris, 1967.
[3] OTTEN (E.-W.), *Naturwissenschaften*, 1964, **51**, 157.
[4] OTTEN (E.-W.), Habilitationsschrift Heidelberg 1966, unpublished.
[5] OTTEN (E.-W.), WINNACKER (A.), *Phys. Letters*, 1966, **23**, 462.
[6] KLUGE (H. J.), OTTEN (E.-W.) and Zimmermann (G), contributed paper to these proceedings, this issue, p. C1-15.
[7] WINNACKER (A.), Diplomarbeit Heidelberg, 1967, unpublished.