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INTRODUCTORY PAPER ON HANLE EFFECT, LEVEL CROSSING AND DOUBLE RESONANCE EXPERIMENTS

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Résumé. — L’auteur passe en revue un certain nombre de techniques expérimentales qui utilisent les propriétés de la diffusion de la lumière par des atomes libres au voisinage d’une fréquence de résonance pour étudier des niveaux atomiques excités :

— L’effet Hanle et ses divers développements récents (effet Hanle moléculaire, effet Hanle en présence d’un champ électrostatique, effet Hanle de niveaux excités par échelons, effet Hanle par fluorescence sensibilisée).
— La double résonance et ses variantes (effets de « battements lumineux », excitation en lumière modulée...).
— Les croisements et anticroisements de niveaux (y compris l’étude de l’effet Stark par croisements de niveaux).

Abstract. — The author reviews experimental techniques which use the properties of the scattering of light by free atoms in the neighborhood of a resonant frequency for investigation of excited atomic energy levels :

— Hanle effect and its recent developments : (Hanle effect in an electrostatic field, molecular Hanle effect, Hanle effect of stepwise excited levels, sensitized fluorescence Hanle effect).
— Double resonance experiments and its variations (« light beats »), excitation with modulated light...).
— Level crossings and anticrossings (including of the study Stark effect by level crossings).

1. Experimental aspects concerning scattering of light on free atoms. — In level-crossing and double resonance experiments photons are scattered on free atoms which are exposed to static as well as alternating electromagnetic fields. These investigations are performed to study the interaction of the light with the atoms, to find the experimental consequences of this interaction and also to deduce spectroscopic data of the atoms involved from the experimental results. Due to the interaction between the radiation and the atoms one photon of the incoming light is absorbed and another photon emitted. With respect to experiments for example the angular distribution of the scattered photon may be studied including measurements of polarisation, or the time dependence of the scattering process investigated. Regarding the excitation of the intermediate state thereby more than one level may be excited coherently by the first photon. Because the scattered photons connect the intermediate state with the final state, the behavior of the state function of the intermediate state during the lifetime is of importance. Taking in mind that the matrix elements contain the eigenfunctions of the atomic states involved a much deeper insight can be gained if changes in the state functions are induced by applying additional perturbations. For example the atoms are investigated in static or alternating electromagnetic fields. In addition second excitations may be studied or the influence of perturbations by collisions investigated. In collision experiments questions of transfer of coherence are of special interest. Because two photons are also involved, if an excited level decays in a cascade through an intermediate state, some effects in resonance scattering of light corresponds to effects in angular correlation.

Regarding the scattering of photons on free atoms the cross-section is normally very small and of the order of the square of the classical electron radius $r_0^2$. However if the energy of the photons corresponds to the energy differences of the states connected by dipol radiation, large cross sections are gained by virtue of resonance scattering. If the atoms would be at rest and the exciting light would have the exact resonance energy, the classical cross section would be of the order of $\lambda^2$. In many experiments the spectral width of the exciting light $\omega$ is larger than the radiation width $\gamma$, and therefore the effective cross section for excitation is decreased by a factor of the order $\gamma/\omega$. For the investigation of excited states which are connected with the ground-state with oscillator-strength between 1 and $10^{-3}$ a
density of about $10^8$-$10^{12}$ atom/ccm is often sufficient with regard to the intensity of the resonance fluorescence. Atoms with high vapour pressure can often been studied in resonance bulbs, while for the investigation of atoms for which a sufficient vapour pressure can be produced only at very high temperature the light is scattered on atomic beams. As a source for the exciting photons low pressure discharges in hollow cathodes are used very often operated with direct current or radio frequency power.

2. Perturbation of resonance scattering of light by static external electromagnetic fields. — In experiments on resonance scattering of light or on cascade decay of excited states like $\gamma\gamma$ angular correlations the initial state is connected with the intermediate state by the radiation field of one photon and again the intermediate state is connected with the final state by the radiation field of the second photon. In the case of resonance scattering of light the first transition corresponds to the absorption of an incoming photon while the second photon is caused by the interaction with the radiation field corresponding to spontaneous emission. In the case of cascade decay both transitions correspond to spontaneous emission.

The probability to observe the photon which is emitted in the second transition depends on the excitation of the intermediate state and on the evolution during the time until the emission process takes place. For the description of the process the axis of quantisation for the atom as well as a direction which specifies the photon is of interest. The scattering process may be regarded for the simple case with an intermediate level with angular momentum $l = 1$, a ground state with $l = 0$ and linear polarisation of the exciting photon. If one chooses for example the axis of quantisation $z$ perpendicular to the polarisation of the exciting light one may take the axis of quantisation $z'$ parallel to the polarisation. Corresponding to the direction $z'$ the linear polarized light connects the ground state $m = 0$ with the excited level $\mu = 0$ of the intermediate state. If one applies a magnetic field again perpendicular to the polarisation of the exciting radiation the eigen functions of the intermediate levels will be mixed due to the magnetic field perturbation during the lifetime of the intermediate state. If the perturbing static field is strong enough that during the lifetime the amplitudes of $\mu = + 1$ and $\mu = - 1$ are getting considerable amounts, the angular distribution of the scattered radiation is changed substantially. This change first detected by Hanle [2] is reached if the magnetic field perturbation $\mu B/h$ will be comparable with the radiation width $\gamma$ of the intermediate state, as it was the case in the first description.

Very similar processes are observed in experiments on perturbed $\gamma\gamma$ angular correlation. Because the multipole character of the photons involved is not only of type $E1$ but may also correspond to electric multipole radiation $E2, E3, ...$ or magnetic multipole radiation $M1, M2, ...$ the situation is of course more general. The probability for observing the photons $k_1$ and $k_2$ after time $t$ is [3]

$$W(k_1, k_2; t) = \sum \left( \frac{m_i m_f}{\mu \mu'} \right) < m_f | H_2 A(t) | \mu > \times$$

$$\times < \mu | H_1 | m_i > < m_f | H_2 A(t) \mu' > \times < \mu' | H_1 | m_i > \times$$

whereby $m_i, \mu$ and $m_f, \mu_f$ designe the magnetic quantum numbers of the initial, intermediate and final state, $H_1$ and $H_2$ are the operators corresponding to the emission of $k_1$ and $k_2$, and

$$A(t) | \mu > = \sum_{m_0} | m_0 > < m_0 | A(t) \mu >$$
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describes the time evolution of the eigen-function in the intermediate state during the lifetime according to the external perturbation. If one does not make measurements which take into account the time $t$ the observations are described by the time integrated probability function and yields as in the case of resonance scattering of light a change of the angular correlation and a decrease of the factor which describes the not isotropic part of the coincidence rate [4]. A substantial change is obtained again if the magnetic field perturbation $\mu H/h$ is of the order of the radiation width, where $\mu$ is the magnetic nuclear moment of the intermediate state. If one distinguishes the two photons with the aid of energy measurement a rotation of the angular $\gamma \gamma$ correlation function is observed [5a]. Using time delayed coincidence technique in $\gamma \gamma$ angular correlation experiments the probability $W(k_1, k_2, t)$ is observed as a function of the delay time and the effect of the time evolution of the intermediate states can be observed directly. For example with fixed positions of the counters and constant magnetic field strength a modulation of the coincidence rate with approximately twice the Lamor frequency of the intermediate state as a function of the delay time is reported [5b]. Measurements are possible for a time interval which amounts a few times of the lifetime of the intermediate state.

In experiments on Hanle effect [2] the perturbation of the resonance scattering of light on free atoms is studied if static magnetic or electric fields are applied which remove the degeneracy of states in zero magnetic and electric fields with angular momentum $J$ or $F$. For the case of experiments in magnetic fields the magnetic field perturbation $\mu g H/m_z$ removes the degeneracy according to the Zeeman effect in first order. If an electric field is applied atomic levels of opposite parity are connected to the state under investigation. The degeneracy of levels with different absolute value of magnetic quantum number $|\mu|$ is removed according to second order perturbation theory corresponding to quadratic Stark-effect. For an electric field parallel to the axis of quantisation the change of the energy of sublevels may be obtained from the perturbation $(\alpha + \beta J_z^2) E_z^2$ [6].

The measurement of the attenuation of the coherent contributions of resonance scattering gives therefore informations for the ratio $g/\gamma$ or $\beta/\gamma$ where $g$ and $\beta$ determine the effect of magnetic and electric fields on the energy levels. Because in many simple atomic spectra the $g$-values of excited levels are well known from theoretical considerations, Hanle effect measurements are used to deduce the radiation width of excited states [7]. In the case where several emission lines, coming from excited levels with different $g_F$ or $g_I$-values, cannot be resolved in the resonance fluorescence further assumptions for the intensities of the excitation process are necessary. If higher density of the atoms investigated are used, effects of coherence narrowing as well as collision phenomena (also for example if additional foreign gases are present) may be of importance. If one applies an electric field [8] simultaneously with a magnetic field the Hanle effect is modified in such a way that the amplitude of signal becomes smaller and the width of the signal larger. From the measured values of $\gamma$ or $\beta$ informations on sums of radial matrix elements of the atom may be obtained and in suitable cases values for oscillator strength can be deduced. Besides measurements on neutral atoms experiments on ions are reported. In connection with the charge of the ions additional care must be taken that the density of the ions is not disturbed by changing the magnetic field [9]. Further experiments on resonance scattering of light on free atoms measurements on molecules are reported showing for example Hanle-effect in an excited state of the diatomic molecule NO [10].

In addition to the investigations of the two photon processes in static external electromagnetic fields further perturbations may be studied. If one is able to get enough atoms in an excited state a second absorption from this intermediate state may be performed corresponding to stepwise excitations and the radiation following second excitation may be observed. For properly chosen polarisations for the excitation and the observation coherence effects of the first or second state may be observed. Experiments are reported involving stepwise excitation of the $3S_1$-state in Cd I-spectrum via the first excited state $3P_1$ [11]. Regarding collision phenomena problems of coherence transfer from one excited atom to another atom by collision are explored with theoretical and experimental investigations. For example in a vapour which consists of a mixture of Hg- and Cd-atoms the levels of the $3P_1$-state of Hg were excited coherently and the observation of coherence processes for the corresponding excited state $5 3P_1$ in Cd I-spectrum is reported [12].

3. Resonance scattering of light in presence of an alternating magnetic field with regard to double resonance experiments. — The application of a magnetic field perpendicular to the axis of quantisation causes a connection of levels with difference in magnetic quantum number $\Delta m = \pm 1$. If the frequency of this
magnetic field does not correspond to the energy difference of the levels involved and if the perturbation caused by this field is small compared with the energy difference of the regarded levels, no considerable admixture of the connected levels can be achieved. But if the frequency of the alternating magnetic field is very near to the energy difference of the regarded sublevels the mixing of the states is very strong, even for small perturbing magnetic fields by virtue of resonance. Magnetic resonance technics are applied in many fields of physics and differ in the detection mechanism of the induced transition. If an alternating magnetic field is applied to an atom, which is in an excited state with different populated magnetic sublevels the mixing of these sublevels during the lifetime may result in a change of the polarisation of the resonance radiation as was first shown by Bitter, Brossel and Kastler [13]. The mixing of states due to the radio frequency field will be considerable if in a rotating frame the corresponding magnetic field perturbation is of the order of the radiation width. With increasing field strength double resonance multiquantum transitions are observed [13a] in similar way as in the atomic beam resonance method. The line width of the double resonance signal is given by the radiation width of the excited state in the limit of vanishing field strength of the perturbing magnetic field and shows for the case of multiple scattering of light coherence narrowing [14]. Besides the change of polarisation due to the radio frequency transitions in the excited state, also changes in the population number of the ground state result and may be used for the detection [15].

Double resonance experiments give informations on absolute values of level distances yielding for example absolute values of hyperfine structure constants or $g_j$-values. Figure 1 shows for example double resonance signals for the transitions between the hyperfinestructure levels $F = 5$ and $F = 4$ of the excited $7^2P_{3/2}$-state in Cs I-spectrum for the isotopes Cs$^{133}$, Cs$^{135}$, Cs$^{137}$ [16]. Investigations of hyperfinestructures for excited states which differ only in the principle quantum number may be used for example to investigate Sternheimer [17] corrections. Many experiments have been performed for energy differences of levels up to several 1 000 Mc/s [17a]. Excited states with lifetime much shorter than $10^{-8}$ s are not easy to investigate, because large magnetic field strength for the radio frequency fields are necessary. Double resonance signals may be used for special detectors for certain isotopes in scanning experiments. If the levels involved belong to states with different parity an alternating electric field perturbation may result in transitions.

With regard to the time dependence of the resonance radiation modulation effects of resonance fluorescence were detected in double resonance experiments by Series [18]. The additional radio frequency field mixes the levels involved coherently. Modulations of the intensity of the resonance fluorescence are reported not only at resonance frequency but also for other frequencies which are deduced from the time evolution of the coherently connected levels. The excitation of resonance fluorescence by modulated light, reported by Corney and Series [18a] provides a further experimental method for studying excited atomic states.
4. Influence of change of coupling of angular momenta on resonance scattering of light. — For atomic states in which finestructure or hyperfinestructure interactions causes finestructure or hyperfinestructure multipletts the application of a sufficient large magnetic field gives rise to a change of the coupling of angular momenta. A substantial change of the eigen-function results if the external magnetic field perturbation becomes of the order of the multiplett splitting in zero magnetic field. The corresponding change of the eigen-functions causes a change of the polarisation degree of the resonance fluorescence. This effect was first used by Heydenburg [19] and co-workers to investigate the strength of the hyperfinestructure interaction of excited states in alkali-spectra. This effect ceases if the coupling for strong magnetic field is reached. But also without external perturbation the polarisation degree of the resonance fluorescence mirrors the coefficients which describe for a certain sublevel of the excited state the contributions of the different orbital angular momentum eigenfunctions with different magnetic quantum numbers. Therefore the value of the polarisation degree gives information on the coupling of spins, if the finestructure or hyperfinestructure interaction is larger than the radiation width.

In the intermediate region between Zeeman-effect and Paschen-Back-effect at certain magnetic field strength the energy of sublevels may become equal. If the difference of the magnetic quantum numbers of these levels are \( \Delta m = 1 \) or \( \Delta m = 2 \) these levels can be excited coherently with electric dipol radiation in the vicinity of the magnetic field strength of the level-crossings. The resulting change in the resonance fluorescence corresponds to the change of the resonance fluorescence due to Hanle-effect in the vicinity of zero magnetic field. These interference signals due to level-crossings in the course of the change of coupling of angular momenta caused by external magnetic field perturbation were first detected by Colegrove, Franken, Lewis and Sands [20] in the excited 2 \( ^3P \)-states of He \( I \)-spectrum. Because the magnetic field strength at which level-crossings occur is determined by the Hamiltonian which contains as parameter the finestructure- or hyperfinestructure interaction constants, and by the strength of the external field perturbation, described by g-values and the magnetic field strength \( H \), the level-crossing technique corresponds to an comparison of these two interactions. Therefore values for the ratio of interaction constants devied by the g-value are deduced from the level-crossing-signals. For example in the case that the finestructure splitting is much larger than the hyperfinestructure, the crossings for hyperfinestructure levels may be calculated from the Hamiltonian [21]

\[
\mathcal{H} = A \mathbf{I} \mathbf{J} + \frac{\alpha}{2} \left[ 3 \mathbf{I}^2 \mathbf{J}^2 + \frac{3}{2} \mathbf{I}^2 \mathbf{J} - \frac{1}{2} \mathbf{J}^2 \mathbf{I}^2 \right] + \\
+ \mu_B g_1 H \mathbf{I} \mathbf{J} + \mu_B g_1 H \mathbf{I} \mathbf{J}.
\]

Because the linewidth is again determined by the radiation width and a factor (of the order of the g-value) which describes the increase of the energy difference of the crossing levels for increasing or decreasing magnetic field strength with regard to the crossing point the level-crossing-signals may provide spectroscopic data with high resolution. The signal width becomes smaller with increasing lifetime, but because long lifetimes are connected with small oscillatorstrength for the transition between the ground state and the excited state, it becomes more difficult to investigate excited states with lifetimes longer than for example \( 10^{-5} \) s by means of resonance scattering of light.

For hyperfinestructure investigations level-crossing-experiments have been performed in the alkali and alkali-like elements for the alkali-earth-elements, trivalent elements and complex spectra [22]. In figure 2 an atomic beam apparatus for level-cros-
sing investigations is shown. Figure 3 shows for example level-crossings in the first excited $4\, p\, ^2P_{3/2}$ state of Cu I-spectrum. If the hyperfine structure interaction is large due to $s$-electrons, the accuracy for the measurement of the magnetic hyperfine structure interaction constant is in suited cases of the order of $10^{-4}$ and better and hyperfine structure anomalies may be determined. If as another extreme the excited state has only a small hyperfine structure interaction which may occur if the hyperfine structure is determined by $p$- and $d$-electrons with small magnetic field at the nucleus, the radiation width is often no more small compared with the hyperfine structure interaction. For this case detailed investigations of the line shape of overlapping level-crossing-signals must be performed. Even for the case that the radiation width is of the same order than the hyperfine structure interaction values for the hyperfine structure constants can be deduced with some accuracy [23].

For level-crossings of sublevels of a finestructure multiplet only the lightest elements can be investigated at the moment, because for other elements the finestructure splitting is too large. In the event of finestructure crossings for isotops with nuclear spin not zero, the additional interaction with the nucleus may give rise to anticrossings which involve the change of eigen-functions near the level-crossing. This effect was first detected by Eck [24] for the first finestructure level-crossing of the $2\, ^2P$ states of Li I-spectrum.

Investigations of the shift of level-crossing field strength caused by the additional application of an electric field yields again information on the Stark-parameter $\beta$, which describes for example for electric field parallel to the magnetic field the electric field interaction proportional to $J_z^2$. The change of the magnetic field strength of the crossing point results if the energy shift of the two sublevels involved are different. For the investigations of the interaction constant $\beta$ the measurement of the shift of a level crossing at $H \neq 0$ may be more suited than the mea-
measurement of the influence of an electric field on the zero field level-crossing, because only two levels are involved. In experiments with an additional electric field the Hamiltonian contains besides the hyperfine-structure interaction and the magnetic field interaction in addition the interaction with the electric field. Therefore from the experimental results a relation between the Stark-parameter $\beta$ and the hyperfine-structure interaction constants may be deduced.

References


