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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 éclatons, 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 γγ 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 r₀. 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 λ³. In many experiments the spectral width of the exciting light ω is larger than the radiation width γ, and therefore the effective cross section for excitation is decreased by a factor of the order γ/ω. 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
be studied in resonance bulbs, while for the investi-
gation of atoms for which a sufficient vapour pres-
sure 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 experi-
ments 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 transi-
tion 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 exci-
tation 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 quanti-
sation 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 quanti-
sation $z$ 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
with the excited level $\mu = 0$ of the interme-
diate state. If one applies a magnetic field again per-
pendicular 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]

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

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

whereby $m_i, \mu$ and $m_f$ designate 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

$$A(t) | \mu > = \sum_{m_b} | m_b > < m_b | A(t) | \mu >$$
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 pro-
bability 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 pertur-
bation \( \mu_I H/h \) is of the order of the radiation width,
where \( \mu_I \) is the magnetic nuclear moment of the interme-
diate state. If one distinguishes the two photons with
the aid of energy measurement a rotation of the angular
\( \gamma \gamma \) correlation function is observed [5 a]. Using time
delayed coincidence technic 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 obser-
ved directly. For example with fixed positions of the
counters and constant magnetic field strength a modu-
lation of the coincidence rate with approximately
twice the Lamor frequency of the intermediate state
as a function of the delay time is reported [5 b]. Mea-
surements are possible for a time interval which
amounts a few times of the lifetime of the interme-
diate 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 magneti-
cic electric fields with angular momentum \( J \) or \( F \). For the case of experiments in magnetic fields the
magnetic field perturbation \( \mu_B g \cdot H \cdot m_z \) 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 inves-
tigation. The degeneracy of levels with different abso-
lute value of magnetic quantum number \( | \mu | \) is removed
according to second order perturbation theory corres-
dponding 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 measure-
ments are used to deduce the radiation width of
excited states [7]. In the case where several emission
lines, coming from excited levels with different \( g \)-
or \( \beta \)-values, cannot be resolved in the resonance
fluorescence further assumptions for the intensities
of the excitation process are necessary. If higher den-
sity 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 scatter-
ing 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 absorp-
tion from this intermediate state may be performed
corresponding to stepwise excitations and the radia-
tion 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 repor-
ted 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 cohe-
rence transfer from one excited atom to another atom
by collision are explored with theoretical and experi-
mental 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 cor-
responding excited state \( 5\,^2P_1 \) in Cd I-spectrum is
reported [12].

3. Resonance scattering of light in presence of an
alternating magnetic field with regard to double reso-
nance 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 techniques 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 multi-quantum transitions are observed [13 a] 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 hyperfine structure 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 hyperfine structures 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 [17 a]. 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 [18 a] 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 multiplets 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 multiplet 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 \( H_i \), the level-crossing technique corresponds to a comparison of these two interactions. Therefore values for the ratio of interaction constants divided 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]

\[
\mathcal{H} = A \mathbf{J} \mathbf{I} + \frac{B [3(\mathbf{I} \mathbf{J})^2 + \frac{3}{2} \mathbf{I} \mathbf{J} - \mathbf{J}^2 \mathbf{I}^2]}{2 J(2 J - 1) J(2 J - 1)} + \mu_B g_1 H Z J Z + \mu_N g_1 H Z I Z.
\]

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 oscillatorenstrength 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-
FIG. 3. — Level scheme and level crossing signals of Cu 65 for the hyperfinestructure of the 4 $^2P_{3/2}$-state in Cu I-spectrum.
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


