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DETERMINATION OF NUCLEAR GROUND STATE PROPERTIES
FAR FROM STABILITY BY OPTICAL PUMPING

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1. Introduction. — This talk is based on experiments [1], [2], [3] performed recently by a visiting team (*) at the ISOLDE facility at CERN with the members: J. Bonn, G. Huber, H.-J. Kluge, U. Köpf, L. Kugler, J. Rodriguez, and E. W. Otten.

The investigation of the hyperfine structure (hfs) and isotopic shift (IS) of optical spectra allows the determination of the basic nuclear ground state properties: spin (I), magnetic moment (μ), electric quadrupole moment (Q₂), and change of the mean squared radius of the nuclear charge distribution between different isotopes (δ < r² >). The subject has therefore widely been explored in the past for stable and long lived isotopes [4], [5] and contributed substantially to the systematic topography of nuclear structure in the valley of stability which could be established within the last two decades. Still basic and quantitative questions are open in understanding nuclear matter. In this situation it makes from first principles. In this situation it makes sense to enlarge the scope of investigations principally.

In particular we should be in a position to choose and change systematically the parameters Z and N of a nucleus independently and free from the boundary condition of being close to the stable valley. With other words, a systematic investigation of nuclear properties along lines of constant Z or N across the full array of nuclei which we can reach in our days, would give us a much wider and more reliable view on the topography of nuclear structure than just the cross section through the stable valley.

Take for example the question of superheavy nuclei. Theoreticians have made detailed predictions about their properties by extrapolating from the knowledge on known nuclei. One should prove whether these theoretical methods lead to reliable statements on (lighter) nuclei far from stability, where experimental material has been made available already now.

2. The ISOLDE facility at CERN. — The ISOLDE at CERN, a collaboration of Danish, French, German, Norwegian and Swedish scientists has been a very successful outliner in the research on nuclei far from stability. The ISOLDE facility [6] is a mass separator set up on line with a hot target (molten metal usually) which is irradiated by 600 MeV protons. Spallation leads to a great number of daughter nuclei centered on the neutron deficient side of the nuclear chart. They have to be separated regarding Z and N before investigation. The separation of the element is chosen by selective evaporation from the hot target. Taking a molten Pb target for instance, Hg is the only volatile daughter element in the neighbourhood. The Hg vapor enters the ion source of a mass separator delivering a chain of about thirty isotopes with a yield reaching 10⁸ nuclei per mass number and second (see Fig. 1). The facility can provide separated isotopes from more than thirty elements. Because of the great number of isotopes the facility usually feeds several experiments simultaneously, most of them attacking their problem by the various tools of ζ, β, γ spectroscopy.

(*) In the course of the experiments the team changed its home university from Heidelberg to Mainz.
3. The optical pumping on line method. — Hfs investigations of isotopes far from stability (or generally of: short lived isotopes) by optical or rf-spectroscopic methods encounter several difficulties. (i) a technical one: the methods have to be adapted to on line conditions. The times for preparation and investigation of the samples shrink down to seconds. (ii) a physical one: the number of atoms produced in saturation is generally too small to apply standard signal detection methods. The sensitivity has to be increased therefore by using the nuclear radiation instead of the photons or rf quanta. This problem has been solved in various elegant ways; for applications in connection with on line separators the atomic beam resonance and the optical pumping method are among the most promising ones (for a discussion see ref. [7], [8]).

The principle idea of Kastler’ method of optical pumping (OP) is to polarize the total angular momentum $F = I + J$ of the atomic ground state by irradiation of circularly polarized resonance radiation, thus transferring the polarized angular momentum of the photons by the process of resonance absorption and reemission (pumping) to the atoms [9], [10]. The detailed mechanism of OP is shown in figure 2 for the Hg atom, the case of our interest where a nuclear spin $I = 1/2$ has been assumed for simplicity. The diamagnetic ground state $1S_0$ has total angular momentum $F = I = 1/2$ because of $J = 0$. It splits into two nuclear Zeeman levels $m_I = +1/2$ and $-1/2$. In the excited $3P_1$ state one obtains two hyperfine components with $F = 1/2, 3/2$ which split into two and four sublevels, respectively. Let us assume that the optical excitation via the resonance line excites only one of the upper $F$ levels, either the $F = 1/2$ or the $F = 3/2$ level. In Hg this is possible in general, since the hfs splitting is large as compared to the Doppler width of the spectral line (compare also figure 4). It is easily shown from the particulars of figure 2 that OP will lead in saturation (neglecting relaxations in the groundstate) to population numbers of the $|m_F >$ substates being proportional to the inverse of the relative absorption probabilities. Thus $<I_z >/I = 100 \%$ and $-50 \%$ are obtained for $F = 1/2$ and $F = 3/2$ pumping resp. Note the role of the buffer gas (noble gas). It causes a complete relaxation of the non spherical excited state within its lifetime of $10^{-7}$ s, but leaves the nucleus in the surrounding of the spherical electronic configuration of the atomic groundstate polarized for seconds. Its main purpose is to prevent too fast relaxations at the paramagnetic walls of the resonance vessel.

For an atomic vapor of reasonable optical thickness the OP effect can be monitored easily by a change in the absorption of the pumping light, since the levels have different relative absorption strength. In the case of a small number of radioactive atoms this may not be detectable, but we have the chance to monitor the polarization (or alignment if $I > 1/2$) by the asymmetry of $\beta$-decay:

$$W_\beta(\beta) = 1 + \frac{\nu}{c} \frac{<I_z >}{I} A \cos \beta,$$

or the $\gamma$-decay anisotropy:

$$W_\gamma(\beta) = \sum_{k \text{ even}} B_k P_k (\cos \beta)$$

($P_k$ are the Legendre polynomials; the coefficients $F_k$ are determined by the degree of nuclear orientation, $B_k$ by the $\gamma$-decay mode, both defined in the literature on angular correlations).
OP experiments based on the $\beta$-detection principle were first performed on some shortlived mirror nuclei in order to determine their moment \cite{11, 12, 13, 14}. A $\gamma$ anisotropy after optical pumping was first observed for the case of $^{203}\text{Hg}$ \cite{15}. The experiments described below are an extension of the methods to isotopes far from stability, delivered by on line mass separators.

The on-line OP set-up is shown in figure 3. The mercury ion beam coming from the separator is stopped on one of the two molybdenum foils. After a collection time of about one half-life, foil I is turned, foil II now collects the ion beam and foil I is heated out at temperatures of above 1 000 °C. Together with a stream of helium buffer gas the mercury atoms enter the quartz vessel. For OP the resonance cell is illuminated by the (6s 6p $^3P_1 - 6s^2$ $^1S_0, \lambda = 2 537$ Å) radiation of a mercury microwave lamp. The circular polarization of the pumping light is periodically alternated by inserting a $\lambda/2$ birefringent quartz plate. Thus, the sign of the nuclear polarization is reversed and any instrumental asymmetry avoided.

$\beta$-radiation containing $^{204}\text{Hg}$, is subjected to a magnetic field in order to match a hyperfine component of the unstable isotope by a Zeeman component of the lamp. Therefore, the decay asymmetry, plotted against the magnetic field, displays directly the hfs. Figure 4 shows the experimental $\beta$-decay asymmetry of $^{185}\text{Hg}$ as a function of scanning magnetic field over the light source. The lamp contains isotopically pure $^{204}\text{Hg}$.

is counted at $0^\circ$, $180^\circ$, and $90^\circ$ in respect of the static magnetic field $H_0$ over the resonance cell. (The $\gamma$-anisotropy is counted by NaI crystals or GeLi diodes under $0^\circ$ and $90^\circ$ with a different polarization scheme allowing the nuclear orientation to flip between $0^\circ$ and $90^\circ$). The nuclear orientation can reach values different from zero only in the case of optical resonance between emission and absorption line. Since (IS) and hfs of this Hg resonance line are bigger than the doppler width of the exciting light, the lamp, containing $^{185}\text{Hg}$, is subjected to a magnetic field in order to match a hyperfine component of the unstable isotope by a Zeeman component of the lamp. Therefore, the decay asymmetry, plotted against the magnetic field, displays directly the hfs. Figure 5 shows the destruction of the $\beta$-decay asymmetry by nuclear resonance of $^{185}\text{Hg}$ as a function of the scanning magnetic field and the thereby deduced level scheme, from which I, $\mu_b$, and IS are determined. In addition $g_1$ can be measured directly by applying NMR to the polarized nuclei while the atoms are in their diamagnetic groundstate, thus destroying the asymmetry (Fig. 5). With a field of a few Gauss over the resonance vessel a satisfying accuracy is obtained already. In favourable cases the asymmetry can be observed directly on the chart recording of the monitor counting rate under $0^\circ$ (Fig. 6).
Figure 7 gives an example for γ-decay anisotropy, observed after pumping the $I = 13/2$ isomer of $^{199}$Hg. Using Ge-Li-diodes anisotropies have been observed in most of the γ-lines of the complex decay schemes of $^{193}$Hg and $^{199}$Hg. It will be proved whether this technique can be used for determination of spins and multipolarities in nuclear decay schemes.

4. Discussion of results. — 4.1. Isotope shift in spherical Hg nuclei. — Figure 8 contains as function of neutron number all hfs- and IS-data, known for this spectral line spanning from $^{181}$Hg to $^{205}$Hg. The stable isotopes are centered around $^{200}$Hg. The results from $^{192}$Hg to $^{203}$Hg including isomers, were obtained before already by standard spectroscopic methods applying off line techniques as far as radioactives were concerned [16]. They include lifetimes down to about one hour which seems to be about the lower limit for experiments of this kind [16], [17]. They established already the widest and most complete set of hfs and IS data collected for the isotopes of one element. This was a very good starting position for our experiments, the results of which are collected in Table I separately.

We will restrict the discussion to the full black points of figure 8 representing the IS. For odd isotopes it is always identical with the shift of the centre of gravity of a hfs multiplet with respect to another isotope. From $^{205}$Hg down to $^{181}$Hg we observe a
null
titative agreement is still lacking. The measurement of a quadrupole moment would be decisive, but unfortunately the nuclear spins are 1/2. A search for rotational bands was negative [21]. The theoretical work suggests a transition from a small oblate to a large prolate deformation for which also the pattern of 0+ energy levels in the lighter neighbours Pt, Os give a hint [22]. Anyhow the situation differs from the one met at the onset of deformed rare earth nuclei. Large zero point motions of the nuclear shape have to be expected, since the potential minima are very flat. They would enter into the IS as well, since the deformation parameter enters into one met at the onset of deformed rare earth nuclei. A second possible explanation is based on Wongs hypothesis of «bubble nuclei» with a hole in the centre [21], [23]. A homogeneously charged bubble with a sharp surface at the inner radius \( R_2 \) and outer radius \( R_1 \) would have approximately the mean squared radius

\[
< r^2 >_\text{b} = < r^2 >_s \left[ 1 + \frac{5}{3} \left( \frac{R_2}{R_1} \right)^3 \right] \tag{4}
\]

where \( < r^2 >_s \) stands for the mean squared radius of a solid sphere with same volume and charge density. Assuming \(^{187}\text{Hg}\) to be a solid sphere and the lighter ones to be bubbles, the observed jump in IS would be fitted by a ratio \( (R_3/R_2) \approx 0.2 \), just the value given by Wong for \(^{174}\text{Yb}\), having the same neutron number as \(^{184}\text{Hg}\). In the bubble model \(^{184}\text{Hg}\) would be a double magic nucleus in the sense that the bubble state reaches its minimum energy as a function of proton and neutron number. There is still no confidence, however, whether this could be the absolute minimum as compared to the normal state and hence represent the ground state.

References