199Hg Mössbauer Measurements on Mercury Alloys and Hg-Fluorides*

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The Mössbauer effect on the 158 keV 5/2−→1/2− transition in 199Hg, of the order of 10 ppm, has been studied using the current integration technique. The isomer shift between the Hg(I)- and Hg(II)-fluorides as well as the quadrupole splitting in Hg2Pt and Hg2F2 are interpreted in terms of relativistic Hartree-Fock-Slater and Molecular Orbital calculations. The following nuclear parameters could be derived:

Δ<r²>=(3.2±1.1) 10⁻³ fm² and Q(5/2−)=(-0.8±0.4) b.

Evidence for an oblate triaxially deformed 199Hg nucleus is derived from particle plus rotor calculations.

1. Introduction

An essential condition for observing a measurable Mössbauer effect (MB) is a sizeable recoilfree fraction or Debye-Waller factor of source and absorber. Due to the exponential dependence on the transition energy a practical limit lies around 150 keV even at low temperatures. The 158 keV transition in 199Hg renders an MB-experiment in particular very difficult. Because of the rather “soft” Hg compounds with low Debye temperatures the resonance effect drops to values of less than 10⁻⁴. A sufficient statistical accuracy is achieved only by collecting large numbers of γ-quanta at very high count rates of >10⁹ c/s for which the common pulse analyzing techniques cannot be employed.

This paper describes an application of the current integration technique at which only part of the information of the detector signal, namely an average of the detector current, is utilized for the determination of the MB-effect.

By means of this technique isomer shifts and quadrupole splittings were observed. Because no atomic data were existing which permitted an analysis of the hyperfine data with regard to the wanted nuclear parameters, relativistic Hartree-Fock-Slater adjusted to Molecular Orbital calculations had to be performed. This is the main content of a second part of this paper.

Finally we will compare the measured quadrupole moment with calculations using the model of a quasiparticle coupled to a triaxially deformed nucleus. For a certain set of chosen parameters excellent agreement with measured multipole moments is achieved. This then supports the picture of a gamma deformed core also for the low lying states.

2. Experiment

The MB-measurements were performed in transmission geometry. Source and absorber were kept at 4.2 K in a helium-bathcryostat. The basic features of the MB setup are shown in Fig. 1. In the current integration technique, the pre- and mainamplifier as well as the single channel analyzer of the single pulse measurement technique are replaced by a voltage to frequency converter (VFC). The pulses from the detector overlap in their collection times if the counting rate is higher than 10⁷ c/s. In this case only an average detector current is measurable. Therefore it is not pos-

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sible to discriminate the energy by a single channel analyzer as in the usual MB setup. A frequency of a pulse train proportional to the detector current is produced by help of a current to voltage (CVC) and a voltage to frequency converter. The effective integration time is given by the distance between two pulses. The pulses are then fed to a multichannel analyzer (MCA) in the multiscaler mode as usually. Further details of the current integration technique have been described in [1] and references cited therein.

The relative velocity between source and absorber was generated by a recently developed electromechanical drive system [1], moving the absorber. Figure 2 shows the arrangement of source and absorber in the cryostat. The effects to be measured are of less than 10^{-4}. Therefore it is essential to reduce the nonlinear geometry effects due to at least this level. This requires that the absorber is moved and in addition multiple scattering is taken into account by choosing carefully the geometrical arrangement depending on the absorber material. Therefore a position monitor was installed in the drive, which allows the determination of the optimum absorber position. This was kept constant by an additional feedback system [1].

The first excited nuclear state in ^{199}\text{Hg} is populated by the decay of the radioactive nucleus ^{199}\text{Au}. The decay scheme is shown in the insert of Fig. 3. The source of about 4 Ci was produced by neutron activation of a 400 mg Pt-metal foil (95.83% ^{198}\text{Pt}) in the FR2 reactor of the KFK Karlsruhe.

In current integration experiments the γ-detector should have a high total efficiency for the MB transition energy and a low one for all other transitions. Furthermore the dynamical response should be linearly dependent on the γ-flux. While NaI-detectors can be adapted to fulfill the first requirement it was found that they have a highly nonlinear behaviour at high doses. With proper choice of the active intrinsic zone a Ge(Li) detector is free of these shortcomings. Care has to be taken with regard to microphonics in the preamplifier, in particular if these are caused by the drive movement, in which case they add coherently to the velocity spectrum. The measurements were performed using a 53 cm³ Ge(Li)-diode with an active volume of about 40 cm³. With the source detector geometry shown in Fig. 2 and with a source of 4 Ci, we achieved count rates up to 10⁹ c/s. A pulse height spectrum of the source is shown in Fig. 3, measured 28 d and 42 d after irradiation, respectively.

3. Analysis of the Mössbauer Spectra

The analysis of the MB spectra has been performed by least square fitting of Lorentzian line shapes to the spectra. In the case of hyperfine split spectra the relative positions and intensities of the hyperfine components were correlated. The Hamiltonian to be used in the presence of an electric hyperfine interaction is in common notation

\[ H_Q = \frac{e Q V_z}{4 I(2I - 1)} \]

\[ (3I_x^2 - I^2)(\eta/2)(I_x^2 + I_z^2) \]  

(3.1)

with

\[ \eta = (V_{xx} - V_{yy})/V_{zz}, \quad 0 \leq \eta \leq 1. \]