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Measurement of the spin dependence of Rayleigh scattering*

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The spin dependence of Rayleigh scattering has been measured by scattering 129-keV γ rays of ^{191}Os on a magnetized iron-cobalt alloy. The circularly polarized γ radiation was produced by nuclear polarization of ^{191}Os alloyed in iron at temperatures of 20–30 mK. For a mean scattering angle of 32° a spin dependence of $\Delta\sigma = (-0.216 \pm 0.033)\%$ has been found, which has the same sign as spin-dependent Compton scattering, the larger cross section appearing for the case where the photon and electron spins are antiparallel.

I. INTRODUCTION

The dependence of the scattering cross section on the relative position of the photon and electron spins is a well-known effect for Compton scattering and used mostly when detecting the circular polarization of the γ radiation. We investigated a similar effect for Rayleigh scattering—the only relevant part of the elastic cross section in the case considered here.

The first hint was given by Daniel and Schmitt,[1] who obtained in a β-(circularly polarized γ) correlation experiment too large a value for the analyzing power ϵ of their magnet if the spin dependence of Compton scattering was taken into account only ($\epsilon = 0.0183 \pm 0.0050$ instead of the calculated value of 0.0046). They proposed to explain this discrepancy by a spin dependence in Rayleigh scattering which, analogous to Compton scattering, may be characterized by

$$\Delta\sigma = (\sigma_+ - \sigma_-)/(\sigma_+ + \sigma_-),$$

where σ_+ and σ_- are the Rayleigh cross sections for parallel and antiparallel alignments of the photon and electron spins. They considered this assumption plausible because bound intermediate states of the scattering electron contribute appreciably, as shown theoretically by Brown and Woodward.[2] Spin dependence of the Rayleigh cross section is possible if the number of available intermediate states of a given spin direction depends on the magnetization direction, as is the case in magnetized iron. From a subsequent Mössbauer and Rayleigh scattering interference experiment, Renz[3] reported a value of $(6.5 \pm 2)\%$ for an energy of 14.4 keV and a scattering angle of 35°. Effects by three orders of magnitude smaller were found by Volk[4] in a transmission experiment with the same energy. Applying also Mössbauer techniques he used Faraday rotation of linearly polarized recoilless γ radiation in ferromagnetic material as detector for the spin dependence.

In a theoretical paper of Brown *et al.*[5] in which the cross section for elastic scattering was calculated, spin dependence was implicitly contained but had no effect on the results because the case of polarized radiation and spin dependence of the occupation of the intermediate states was not considered. This spin dependence was also neglected by Jähnig[6] in his calculations performed for Fe in the form-factor approximation. Consequently, the resulting values were two orders of magnitude too small to explain the experiments.[1,3] Jähnig[7] was, however, able to show an asymmetry in the calculated Mössbauer line structure which can explain the asymmetry reported by Renz, and attributed by him to be due to a Mössbauer-Rayleigh interference. In order to clarify the situation, we decided to perform a new experiment with improved technique, to enable a clear-cut interpretation of the result.

In the present experiment nuclear polarization of ^{191}Os at temperatures of 20–50 mK was used to obtain 129-keV γ radiation with a circular polarization of 98–100% measured in the direction of the magnetic field. The photons were scattered outside of the cryostat on magnetized iron. In order to vary the scattering angle (and also to vary the stray field, cf. Sec. IV) three magnets with different geometries were used. Two Ge(Li) detectors with fairly high resolution were used to separate Rayleigh from Compton scattering. The relative counting difference, $\delta = (N_+ - N_-)/(N_+ + N_-)$, where "plus" and "minus" define the direction of magnetization in the scatterer, was used to extract the spin dependence.

II. EXPERIMENTAL SET-UP

As source of the polarized γ radiation ^{191}Os was chosen mainly because of its large polarizability even at relatively high temperatures (50 mK) and its dominant γ line at 129 keV, which is low enough in energy for the investigation of Rayleigh scattering and, moreover, fairly close to the energy of the first experiment.[1] The absence of stronger lines avoids difficulties from a spin-dependent Compton background. The decay scheme of ^{191}Os is shown in Fig. 1.

Inactive ^{190}Os (enriched to 95.5%) was implanted in iron. A sample with a concentration of 9.8-at.% ^{190}Os in iron was irradiated for 14 days in the Munich Research Reactor with a neutron flux of 10^{13} neutrons sec^{-1} cm^{-2}. The experimental set-up is shown in Fig. 2. The sample with an initial activity of 6.3 mCi was soldered by Wood's metal on a copper sinter block fixed in the mixing chamber of the dilution refrigerator. Sample temperatures varied from 50 to 20 mK depending on the radioactive heating. During the main runs the refrigerator was operated at lowest temperature for three months without interruption.

The sample was magnetized by a 2.5 kG split-coil superconducting magnet, and the ^{191}Os polarized by the strong hyperfine fields in the iron. Saturation was already achieved at 1 kG. The radiation emitted by the source passed through five cylindrical aluminum walls (shields and vessels for nitrogen and helium with a total thickness of 4 mm). One of three different scattering magnets was used as scatterer, two magnets with cylindrical geometry[8,9] and inner diameters of 10 cm (Fig. 2) and 14 cm, respectively, and a 100°-segment magnet with an inner diameter of 14 cm of the corresponding cylinder.[9] Two conical lead shields with different shapes for each of the three magnets determined the angle of incoming and scattered radiation and thus the scattering angle. A third lead shield absorbed the direct

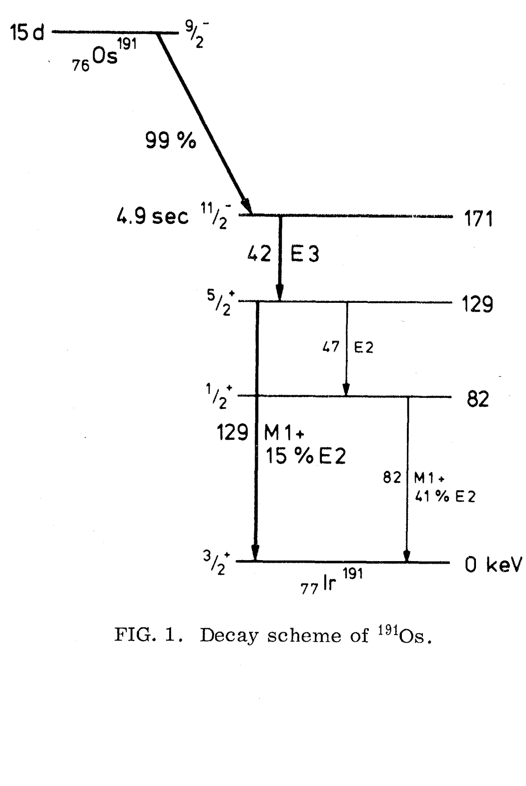

FIG. 1. Decay scheme of ^{191}Os.

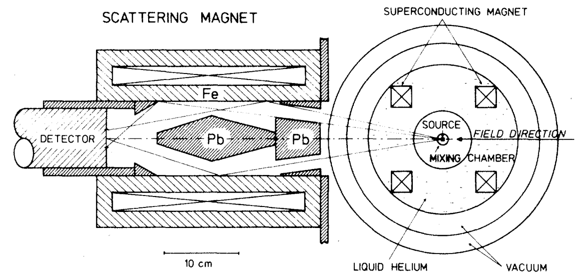

FIG. 2. Scattering set-up.

radiation. For the measurements with mean scattering angles of 32° (main measurement), 40°, and 55° one of the two cylinder magnets and a planar Ge(Li) detector (Canberra) with an active area of 9 cm^2, an efficiency of 80% and a resolution (FWHM) of 900 eV at 129 keV were used. The measurements with a mean scattering angle of 21°, 29°, and 35° were performed with the segment magnet, partly simultaneously in 180° position to the other magnet, and a true-coaxial Ge(Li) detector (Canberra) with an active area of 6 cm^2, an efficiency of 60% and a resolution of 900 eV at 129 keV.

The detector pulses were amplified and stored in one of two 1k divisions of a Nuclear Data 50-50 System depending on the magnetization direction. After an accumulation time of 200 sec for one direction of magnetization the magnetization was automatically reversed, stopping the measurement for 10 sec, and accumulation started again. After a measuring time of two hours the data were read out on magnetic tape.

In order to monitor the sample temperature a second Ge(Li) detector in 180° position to the first was used in most runs to measure the intensity of the unscattered radiation. The 129-keV peak was analyzed during the measuring pause for reversing the field and its intensity stored in another part of the memory to monitor temperature variations.

III. POLARIZATION OF ^{191}Os

In order to calculate the degree P_c of circular polarization for the 129-keV γ radiation one must know the population of the hyperfine substates of the $\frac{11}{2}$-excited state of ^{191}Ir, which depends on the ratio of the spin-lattice relaxation time to its lifetime. Nuclear orientation measurements by Hagn[10] yielded a spin-lattice relaxation time of the order of the lifetime. So both the ground-state polarization of ^{191}Os and the spin-lattice interaction of the $\frac{11}{2}$ state of ^{191}Ir determine P_c. From the anisotropy data a temperature-dependent correction factor $f(T)$ for the splitting constant μH was calculated so that the corrected "splitting constant" $f(T)\mu_{Ir}H_{Ir}$ yields the observed anisotropy. P_c was then obtained using this corrected splitting constant and formulas by Tolhoek *et al.*,[11] namely the expressions (1)-(3), (6), and (19) of his paper, assuming an $E2/M1$ mixing ratio[12] of -0.39.

As can be seen from Fig. 3, the difference in polarization between the $\frac{11}{2}$ state of Ir and the 129-keV state is smaller than 2% between 20 and 50 mK (range of the measurements).

IV. SYSTEMATIC ERRORS AND CONTROL MEASUREMENTS

For measurements with a relative difference in counting rates of a tenth of a percent it is indispensable to know the magnitude of possible systematic errors, particularly of those errors which are due to the change of magnetization of the scatterer. In the following we want to discuss these possible errors.

The relative difference in counting rates caused by the decay of the source was less than 5×10^{-5} under our conditions, and hence could be neglected compared to the ten times larger statistical error.

Heating effects of the source by eddy currents induced by the stray field of the scattering magnet during change of magnetization could cause a change in anisotropy but were not observed in a separate test experiment. In this experiment the magnetization was reversed every 10 sec for one hour and the unscattered radiation was recorded separately for the two directions of magnetization. The measured effect was $\delta=(0.02\pm0.06)\%$.

The dependence of the detection efficiency on the direction of the stray field was investigated for both detectors and detector positions of 0° and 30° with respect to the axis of symmetry. The results given in Table I indicate no dependence on the direction of the magnetic field.

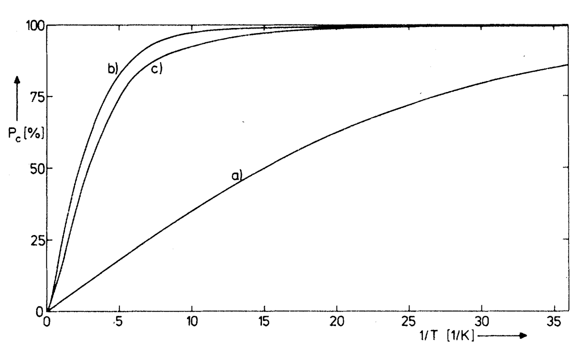

FIG. 3. Temperature dependence of the degree of circular polarization for the 129-keV transition of ^{191}Os. (a) Calculated from the initial population of the ^{191}Os ground state only. (b) Calculated from the initial population of the $\frac{11}{2}$ state of ^{191}Ir only. (c) Actual values calculated from the measured anisotropy data of Ref. 10.

Electronic asymmetries were tested by feeding a test pulse to the preamplifier test input of the detector and found to be negligible $[\delta=(0.005\pm0.010)\%]$.

The change of attractive and repelling forces acting between polarizing and scattering magnet could change the source position and thus cause different counting rates by changing the solid angle. To check this effect, two different kinds of null measurements were performed, one with warm source (4 K) and the same set-up as in the main experiment, and another with cold source but without the lead shield against direct radiation. The results were $\delta=(-0.03\pm0.05)\%$ and $\delta=(+0.022\pm0.018)\%$ at 4 K and 40 mK, respectively. They showed no significant deviation from zero but were used to correct the result of the final experiment.

V. MAIN EXPERIMENT AND RESULTS

In the main experiment the spin dependence of Rayleigh scattering was measured for an energy of 129 keV and a scattering angle of 32°. Supplementary runs were performed for scattering angles of 21°, 29°, 35°, 40°, and 55°. Simultaneously the spin dependence of Compton scattering was measured for the corresponding scattering angles.

The spectrum from the main experiment is shown in Fig. 4(a). This spectrum is the sum of the two scattering spectra for the two magnetization directions of the scatterer. The Pb x rays produced in the lead shield reach the detector via Rayleigh scattering, and so do the Os x rays produced in the source. The test pulse line at about 140 keV was used to check the electronic equipment during the run. In Fig. 4(b) δ is plotted. The sign of δ is positive when the cross section with parallel spins of photon and electron polarized in the iron is larger than with antiparallel spins.

TABLE I. Asymmetry of the detector efficiency for the two used Ge(Li) diodes and two angles of the detectors to the axis of symmetry.

Detector	Angle	δ (%)
Planar	0	$+0.01\pm0.03$
	30°	$+0.02\pm0.03$
"True-coaxial"	0	$+0.01\pm0.02$
	30°	$+0.01\pm0.03$

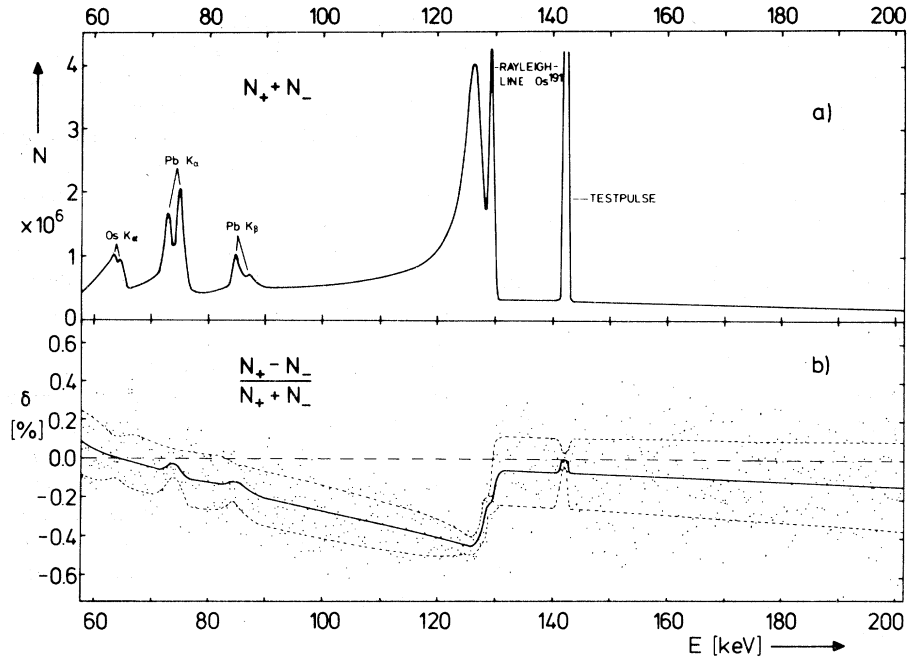

FIG. 4. (a) Total scattering spectrum of ^{191}Os for a scattering angle of 32° (sum of the two spectra measured for the two directions of magnetization of the scattering magnet). (b) Calculated anisotropy [same abscissa scale as (a)]. Dashed lines: value ± standard deviation.

Spectrum and δ vs energy for the Rayleigh line of 129 keV and the Compton continuum below are shown in more detail in Fig. 5. A slight deviation from $\delta = 0$ in the Rayleigh portion can be recognized but a definite statement cannot be given before the effect of the background is carefully examined. There are two components of the background affecting the δ plot in the Rayleigh portion, the low-energy portion of the spectrum from Compton-scattered photons of higher energy, mainly from ^{59}Fe (1095 and 1292 keV), ^{185}Os (646 keV) and ^{192}Ir (296, 308, 317, and 468 keV) and the high-energy slope of the Compton continuum from the 129-keV line of ^{191}Os which extends under the experimental Rayleigh peak. The radiation from ^{192}Ir is partially circularly polarized

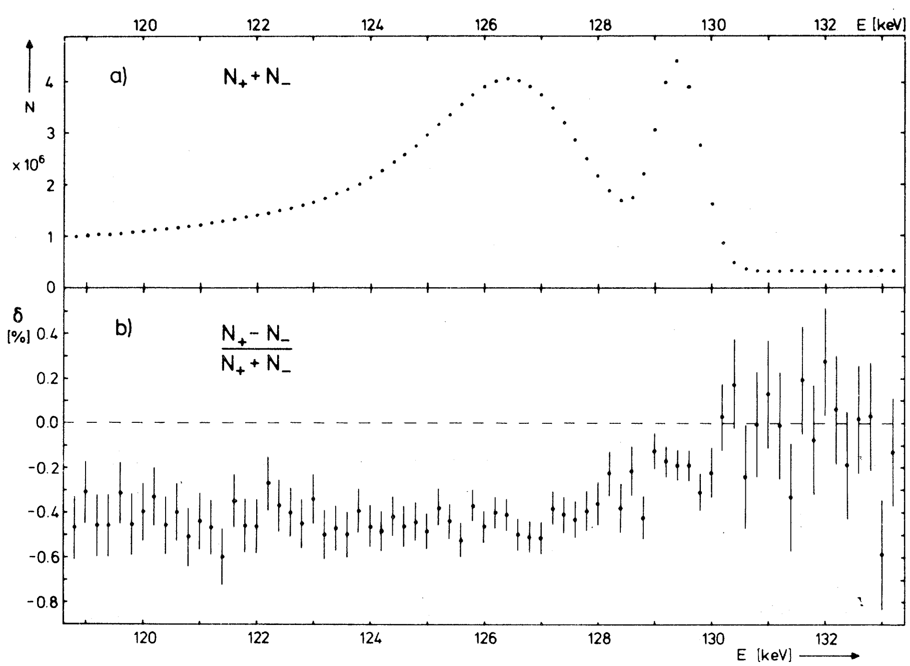

FIG. 5. (a) Region of Rayleigh and single Compton scattering of the total scattering spectrum shown in Fig. 4(a). (b) Calculated anisotropy in the Rayleigh and Compton scattering regions [same abscissa scale as (a)].

(between 40% and 80% for the different transitions) and can cause a counting difference in the background. This effect was corrected for by extrapolating the background linearly for each of the two spectra obtained at the two magnetization directions and subtracting the corresponding intensities separately. The background originating from the Compton slope of ^{191}Os was calculated by fitting two separate Gaussian curves to the slopes of the two spectra corresponding to the two magnetization directions and extrapolating them under the Rayleigh peaks. The calculated intensities were subtracted, and two symmetric Rayleigh lines without any background were obtained. This correction method is shown graphically for one spectrum in Fig. 6.

The values of the full width at half maximum of the summed-up Rayleigh line amounted to 950 eV, compared to a purely instrumental width of 900 eV measured under ideal conditions. Values of δ were calculated for different regions symmetric around the Rayleigh line maximum to find the value with the maximum information, i.e. smallest error, and also to test the effect of the background. If, for example, δ is calculated for a very small energy range only information is lost because the number of events registered in this range is small. If, on the other hand, a very broad energy range is admitted, events far away from the Rayleigh peak are included which contain no information on the Rayleigh scattering but increase the statistical error; portions to the left of the peak are, moreover, the more affected by the Compton slope subtraction the more the energy decreases.

The individual values of δ obtained during the various steps of correction for the different peak regions are tabulated in Table II. The variation of the values of δ with all corrections is not larger than the total error, which demonstrates the insensitivity of the result on the performed corrections.

For the region with the smallest total error, that is channel 637–643 (128.7–130.1 keV), one obtains $\delta = (-0.200 \pm 0.029)\%$ and after correction for systematic errors, $\delta = (-0.211 \pm 0.033)\%$.

Because of the nonvanishing temperature and, to a larger extent, because of the finite opening angle of the scattering magnet the circular polarization of the γ radiation is smaller than 100%. In order to calculate the δ value for scattering of completely circularly polarized radiation the experimental value was correspondingly corrected. One obtains for the corrected δ value, the spin dependence of Rayleigh scattering,

$$\Delta\sigma = (-0.216 \pm 0.033)\% .$$

The results for δ at other scattering angles which were obtained by the same method, are listed in Table III. They are consistent with the result of the main measurement but the statistics are not sufficient to allow any statement on an angular dependence of the spin dependence.

As additional tests of the consistency of the results of spin dependence of Rayleigh scattering, the spin dependence of the Compton scattering recorded simultaneously in the scattering experi-

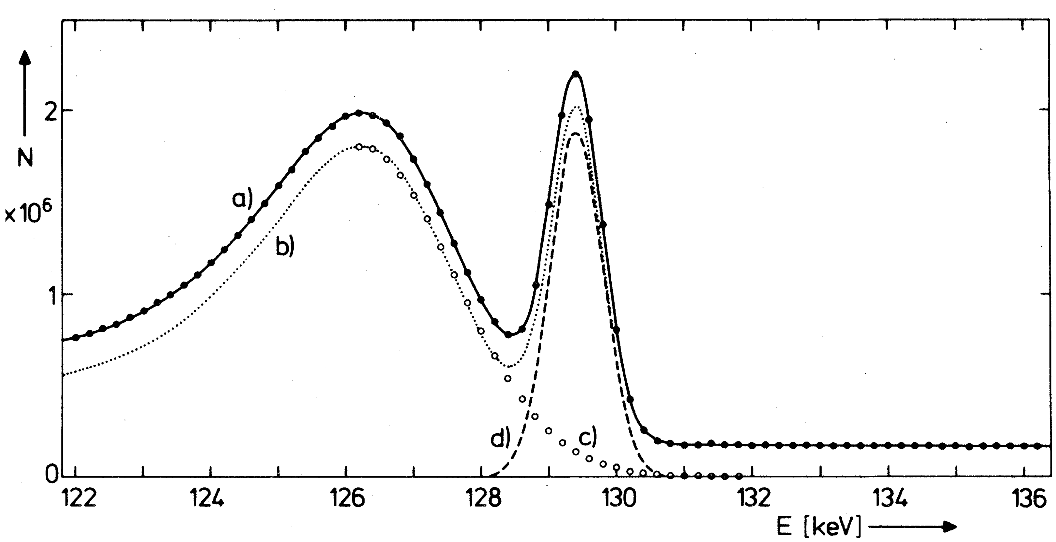

FIG. 6. Illustration of the correction method for background elimination. (a) Uncorrected spectrum. (b) Spectrum after elimination of linear background. (c) Fit of a Gaussian curve to the Compton slope. (d) Spectrum after elimination of both linear and nonlinear background.

TABLE II. Asymmetry for different regions of the Rayleigh peak (columns) and for the different steps of the correction process (rows). (a) Uncorrected counts and calculated asymmetry. (b) Counts and asymmetry of the linear background. (c) Resultant counts and asymmetry after subtraction of the linear background. (d) Counts and asymmetry of the nonlinear background. (e) Resultant counts and asymmetry after subtraction of linear and nonlinear background.

Channel		634–646	635–645	636–644	637–643	638–642	639–641	640
Energy range (keV)		128.2–130.6	128.4–130.4	128.6–130.2	128.8–130.0	129.0–129.8	129.2–129.6	129.4
(a)	N_+	14 452 602	13 329 769	12 231 596	10 925 966	9 022 094	6 120 588	2 189 856
	N_-	14 514 367	13 386 434	12 282 695	10 973 501	9 056 689	6 142 937	2 198 050
	δ (%)	−0.213 ± 0.019	−0.212 ± 0.019	−0.208 ± 0.020	−0.217 ± 0.021	−0.191 ± 0.024	−0.182 ± 0.029	−0.187 ± 0.048
(b)	Fraction	8.2%	7.6%	6.7%	5.9%	5.1%	4.5%	4.2%
	N_+	1 192 896	1 009 374	825 852	642 329	458 806	275 283	91 761
	N_-	1 195 226	1 011 345	827 464	643 583	459 702	275 821	91 940
	δ (%)	−0.10 ± 0.06	−0.10 ± 0.07	−0.10 ± 0.08	−0.10 ± 0.09	−0.10 ± 0.10	−0.10 ± 0.13	−0.10 ± 0.23
(c)	N_+	13 259 706	12 320 395	11 405 744	10 283 637	8 563 288	5 845 305	2 098 095
	N_-	13 319 141	12 375 089	11 455 231	10 329 918	8 596 987	5 867 116	2 106 110
	δ (%)	−0.224 ± 0.021	−0.221 ± 0.022	−0.216 ± 0.022	−0.225 ± 0.023	−0.196 ± 0.025	−0.186 ± 0.031	−0.191 ± 0.051
(d)	Fraction	26.0%	21.7%	17.7%	14.2%	11.5%	9.7%	8.8%
	N_+	3 747 088	2 883 884	2 158 122	1 550 385	1 037 659	594 394	193 402
	N_-	3 775 363	2 905 331	2 173 938	1 561 590	1 045 068	598 599	194 764
	δ (%)	−0.38 ± 0.04	−0.37 ± 0.04	−0.37 ± 0.05	−0.36 ± 0.06	−0.36 ± 0.07	−0.35 ± 0.09	−0.35 ± 0.16
(e)	N_+	9 512 618	9 436 511	9 247 622	8 733 252	7 525 629	5 250 911	1 904 693
	N_-	9 643 778	9 469 758	9 281 293	8 768 328	7 551 919	5 268 517	1 911 346
	δ (%)	−0.164 ± 0.033	−0.176 ± 0.031	−0.182 ± 0.030	−0.200 ± 0.029	−0.174 ± 0.030	−0.167 ± 0.036	−0.174 ± 0.058

TABLE III. Spin dependence of Rayleigh scattering for different scattering angles (including the result of the main measurement). The symbols in column 2 mean (a) segmented magnet, (b) cylindrical magnet with an inner diameter of 10 cm, and (c) cylindrical magnet with an inner diameter of 14 cm.

Mean Rayleigh scattering angle	Scattering magnet	Source temperature (mK)	Degree of circular polarization [a] (%)	Spin dependence $\Delta\sigma$ (%)
21°	a	50	98	-0.27 ± 0.20
29°	a	36	99.5	-0.10 ± 0.19
32°	b	37	99.5	-0.22 ± 0.03
35°	a	39	99.5	-0.21 ± 0.15
40°	c	31	99.5	-0.24 ± 0.12
55°	c	21.5	100	-0.05 ± 0.12

[a] Along the field distribution.

ments and the intensity ratio between Rayleigh and Compton scattering of the 129 keV γ radiation were determined and compared with calculated values.

In order to obtain the spin dependence of the Compton cross section, δ was determined from the region of single scattering as limited by the energies corresponding to the largest and smallest scattering angles possible. The corrections for background and systematic errors were performed in the same way as described for Rayleigh scattering except, of course, for the Compton slope correction. The mean scattering angles for Compton scattering differ slightly from those for Rayleigh scattering because of the different angle dependence of the cross section.

In order to be able to compare the results with theoretical values one has to calculate[13] the analyzing power of the various magnets used. These calculations took into account the different geometries of the scattering magnets, the energy- and angle-dependent detector efficiency and the angular dependence of intensity and circular polarization of the radiation emitted by the source. The agreement of measured and calculated $\Delta\sigma$ values listed in Table IV is good.

Further the measured intensity ratios of Rayleigh and Compton scattering were compared with calculated values which were obtained with a formula by Moon[14] for the Rayleigh scattering cross section and with the Klein-Nishina formula[13] modified by the incoherent scattering function, which takes the effect of bound electrons on Compton scattering[15] into account. This modification reduces the Compton cross section by (2–3)% where the effect is increasing with decreasing scattering angle. The measured and calculated intensity ratios are listed in Table IV and are in good agreement.

VI. DISCUSSION

The measurements described in the present paper prove the existence of a spin dependence of Rayleigh scattering. A clear-cut result was

TABLE IV. Comparison between calculated and measured values $\Delta\sigma_c$ for spin-dependent Compton scattering and between calculated and measured intensity ratios $(R/C)_{\rm theor}$ and $(R/C)_{\rm expt}$ of Rayleigh and Compton scattering, respectively, for the six measured scattering angles.

Mean Compton scattering angle	Spin dependence $\Delta\sigma_c$ $(\Delta\sigma_c)_{\rm theor}$ (%)	$(\Delta\sigma_c)_{\rm expt}$ (%)	Intensity ratios $(R/C)_{\rm theor}$	$(R/C)_{\rm expt}$
22°	-0.19	-0.20 ± 0.11	0.74	0.66 ± 0.05
33°	-0.33	-0.21 ± 0.11	0.32	0.28 ± 0.03
36°	-0.38	-0.39 ± 0.05	0.24	0.23 ± 0.02
41°	-0.48	-0.51 ± 0.08	0.16	0.15 ± 0.03
42°	-0.48	-0.46 ± 0.10	0.13	0.12 ± 0.02
62°	-0.76	-0.64 ± 0.12	0.06	0.04 ± 0.01

achieved for an energy of 129 keV and a mean scattering angle of 32°. At other angles consistent results were obtained which, however, considered isolated, would not give definite answers due to lack of statistics.

The magnitude of the spin dependence of Rayleigh scattering necessary to fully explain the observed ϵ value of Daniel and Schmitt[1] (cf. Sec. I) would be by far larger than suggested by the present experiment. There is, however, no discrepancy because the statistical error in the first experiment[1] is large and, moreover, the experimental conditions differed. Daniel and Schmitt chose a γ-ray energy of 81 keV and a mean scattering angle of 53°. The two experiments cannot directly be compared because of a lack of a theory.

Also because of this lack of a theory, no direct comparison with the experiments of Renz and Volk, and between these experiments, is possible. Among all the earlier experimenters[1,3,4] Renz only claimed to have demonstrated the existence of a spin dependence of Rayleigh scattering, a statement not justified in the light of Jähnig's thesis.[7] Thus the present experiment is, to our knowledge, the first one to establish this effect.

The only author to claim to have calculated the spin dependence of Rayleigh scattering is Jähnig. However, his treatment is, in our opinion, not adequate since he suppressed the main effect from the beginning on (cf. Sec. I). Hence we believe no adequate theory exists. We hope that our investigation will stimulate a thorough theoretical treatment. We expect that physics will benefit threefold from such a calculation: a new method of testing quantum electrodynamics, new insight in the electronic structure of ferromagnetic material, and, perhaps, use of spin-dependent Rayleigh scattering as a tool, for example in measuring the circular polarization of quantum radiation.

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