

HYPERFINE BROADENING OF THE X-RAY LINES

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Results of experiments on the observation of hyperfine broadening of K_{α_1} x-ray lines (HFI effect) due to the interaction between the nuclear dipole magnetic moment and electron current of the atomic shell with a vacancy in the $1s_{1/2}$ state are reported. The experiments were performed with Sb^{121} and Eu^{151} . The theoretical values of the effects are considered. The experimental and theoretical values are in agreement within the experimental errors. It is shown that broadening of the L_{II} level of Np^{237} previously observed by Merrill and DuMond^[8] cannot be ascribed to the HFI effect; this removes the discrepancy between theory and experiment mentioned by the authors. Along with HFI broadening, the values of the isotopic shifts of the K_{α_1} lines for the isotope pairs $Eu^{151}-Eu^{153}$ and $Sb^{121}-Sb^{123}$ are measured. The corresponding changes in the charge radii of the nuclei are discussed.

INTRODUCTION

THE effect of magnetic hyperfine interaction (HFI), which leads to a hyperfine structure of emission lines, is well known in optical spectra of ordinary atoms^[1]. It is observed also in the x-ray spectra of μ -mesic atoms (cf, e.g.,^[2]). The HFI effect is described by the interaction of the magnetic dipole moment of a nucleus with the current of the electronic (mesic) shell of the atom. It leads to a splitting of the fine-structure terms of the atom into sublevels making up the hyperfine structure. The number of components is determined by the number of possible values of the total angular momentum of the atom. The emission lines are likewise split accordingly. In the x-ray spectra of ordinary atoms, however, the HFI effect, which was theoretically predicted by Breit back in 1930^[3], could not be observed experimentally, in spite of numerous attempts^[4-7]. The reason is that the distances between the components of the hyperfine structure are small in comparison with the natural widths of the x-ray lines. At such a ratio of the width and the splitting, the HFI effect leads only to a broadening of the lines (see the Appendix). The value of the broadening is at best $10^{-3}-10^{-4}$ of the line width.

Merrill and DuMond^[8] reported in 1951 observation of the HFI effect in the L-series x-ray lines of the Np^{237} nucleus. From the broadening of the Np^{237} L_{II} level, they have concluded that the experimental value of the splitting is double the theoretical value. As shown at the end of the present section, this conclusion is in error, and the observed broadening of the Np^{237} L_{II} level cannot be the result of the HFI.

The present study was undertaken with an aim at experimentally observing the HFI effect in x-ray lines. We use the following notation: $-e$ and m_0 are respectively the electron charge and mass, $\hbar = h/2\pi$, where h is Planck's constant, c is the speed of light, α is the fine-structure constant ($\alpha = e^2/\hbar c$), E_0 is the electron rest mass ($E_0 = m_0 c^2$), a_0 is the first Bohr radius, μ is the nuclear magnetic moment, m_p is the proton mass, Ze is the charge of the nucleus, A is the mass number,

R is the radius of the nucleus, μ_N is the nuclear magneton, I is the nuclear spin operator, j is the electron total angular momentum operator, F is the atom total angular momentum operator (the quantum numbers corresponding to these operators will be denoted by the same letters I , j , and F), μ is the magnetic dipole moment operator of the nucleus, α is the Dirac four-matrix, $\gamma_1 = [1 - (\alpha Z)^2]^{1/2}$, $\gamma_2 = [4 - (\alpha Z)^2]^{1/2}$.

THEORY

The HFI effect for electrons of x-ray levels was first considered by Breit^[3]. The problem was solved by perturbation theory. The electronic parts of the unperturbed wave functions were the exact solutions of the Dirac equation for the electron in the Coulomb field of the nucleus. By diagonalizing the corresponding matrices of the operator $e\alpha[\mu \times r]/r^3$, which describes the interaction of the field of a point-like magnetic dipole moment with the electron-shell current, he determined the values of the hyperfine splittings for the levels $1s_{1/2}$ and $2s_{1/2}$ in first-order of perturbation theory.

By calculations analogous to those of Breit^[3], we found that the shift of a hyperfine-structure sublevel with total angular momentum F from the unperturbed level (in the case of the levels $ns_{1/2}$, $np_{1/2}$ and $np_{3/2}$, which is more general than Breit's case), can be described by the expression

$$\Delta^F = \pm e\mu_N \mu \frac{F(F+1) - I(I+1) - j(j+1)}{Ij(j+1)} \int_0^\infty fg dr. \quad (1)$$

The plus sign pertains to the $np_{1/2}$ levels and the minus sign to the $ns_{1/2}$ and $np_{3/2}$ levels; f and g are Dirac radial wave functions for the electron in a centrally-symmetrical field with a scalar potential $\varphi(r)$. The maximum distance between the sublevels of the hyperfine structure (hyperfine splitting) is

$$\Delta = |\Delta^{F=I+j} - \Delta^{F=|I-j||}. \quad (2)$$

For a Coulomb potential ($\varphi(r) = Ze/r$) we obtain

$$\Delta^{(A)}(1s_{1/2}) = G \frac{2(\alpha Z)^3}{3\gamma_1(2\gamma_1 - 1)}, \quad (3)$$

$$\Delta^{(k)}(2s_{1/2}, 2p_{3/2}) = G \frac{(\alpha Z)^3}{3\sqrt{2}} \frac{(2\gamma_1 + 2)^{1/2} \pm 1}{\gamma_1(4\gamma_1^2 - 1)(1 + \gamma_1)^{1/2}}, \quad (4)$$

$$\Delta^{(k)}(2p_{1/2}) = G \frac{(\alpha Z)^3}{40\gamma_2(4\gamma_2 - 1)}, \quad (5)$$

where $G = \alpha E_{0\mu} N_{\mu} (2I + 1)/I$. In the numerator of the last fraction in (4), the plus sign corresponds to the state $2s_{1/2}$ and the minus sign to $2p_{1/2}$. Equation (5) has been written out for the case $I \geq j$. Formulas (3) and (4) (for the states $1s_{1/2}$ and $2s_{1/2}$) coincide with Breit's results^[3].

In the expressions (3)–(5) it is necessary to introduce corrections for the finite dimensions of the nucleus (allowance for the redistribution of the electric-charge density (ϵ_e) and of the magnetization density (ϵ_m) over the volume of the nucleus), and also for the screening of the field of the nucleus by the other electrons (η). Later on, in the comparison of the theory with experiment, we shall need only the splitting of the $1s_{1/2}$ level, since the experimentally-investigated K_{α_1} line corresponds to the transition of an electron from the $2p_{3/2}$ level to the $1s_{1/2}$ level, but $\Delta^{(k)}(2p_{3/2}) \approx \Delta^{(k)}(1s_{1/2})/100$, and the splitting of the $2p_{3/2}$ level can be neglected. Taking the corrections into account, the expression for the hyperfine splitting of the $1s_{1/2}$ level can be written in the form

$$\Delta(1s_{1/2}) = \Delta^{(k)}(1s_{1/2})[1 - (\epsilon_e + \epsilon_m + \eta)]. \quad (6)$$

All three corrections decrease the effect and add up to approximately 0.1. We calculated the correction ϵ_e (which is the largest of the three) by using in (1) the wave functions of the $1s_{1/2}$ state of the electron in the field of a nucleus with a uniform distribution of the charge over the volume (Babushkin^[9]). With sufficiently good approximation, we have

$$\epsilon_e = \left(\frac{2ZR}{a_0}\right)^{2\gamma_1-1} \left[\frac{1}{\Gamma(2\gamma_1)} + 2(2\gamma_1 - 1)\Delta n \exp\left(-\frac{2ZR}{a_0}\right) \right], \quad (7)$$

where

$$\Delta n = \frac{L - \epsilon_0 M}{\Gamma(2\gamma_1) [(1 - \gamma_1) - \epsilon_0 M (1 + \gamma_1)]},$$

$$\epsilon_0 = (1 - \gamma_1^2)^{1/2} (1 + \gamma_1)^{-1}$$

The correction ϵ_m ($\approx 1\%$) was determined from data of^[1] for the case of a uniform distribution of the magnetization density over the nucleus.

Since the $1s_{1/2}$ shell is the innermost in the atom, the corrections for screening are small ($\approx 2\%$) and were estimated from Wilson's data^[10] on the ratio of the square of the wave function (at $r = 0$), calculated by the Hartree-Fock method, to the corresponding value obtained from a solution of the Schrödinger equation for one electron in a Coulomb field. Allowance for screening by Slater's method^[11] yields approximately the same results¹⁾. A comparison of the theoretical values of the splittings of the K level ($1s_{1/2}$ state) of Sb^{121} and Eu^{151} with experiment will be presented in the next section.

¹⁾It should be noted that the possible influence of the electric quadrupole splitting of the $2p_{3/2}$ level on the investigated effect is small. Estimates based on the formulas for the optical terms [1] give values $<1\%$ of the magnetic splittings of the state $1s_{1/2}$.

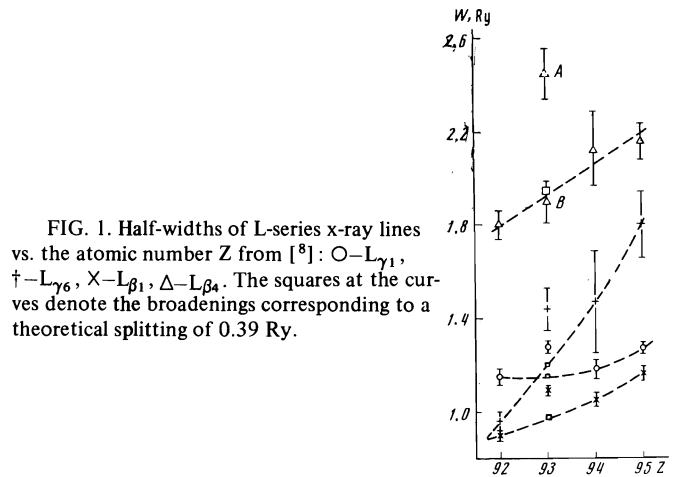


FIG. 1. Half-widths of L-series x-ray lines vs. the atomic number Z from [8]: \circ — L_{γ_1} , \dagger — L_{γ_6} , \times — L_{β_1} , Δ — L_{β_4} . The squares at the curves denote the broadenings corresponding to a theoretical splitting of 0.39 Ry.

For the L-shell ($2p_{1/2}$ state) we attempted to compare the results with the data of Merill and DuMond^[8]. They proposed to observe the HFI effect by investigating the Z-dependence of the widths of the lines $L_{\beta}(M_{IV} \rightarrow L_{II})$, $L_{\gamma_1}(N_{IV} \rightarrow L_{II})$ and $L_{\gamma_6}(O_{IV} \rightarrow L_{II})$. Since the magnetic moment of the Np^{237} greatly exceeds the magnetic moments of U^{238} , Pu^{239} and Am^{241} , the widths of these lines in Np^{237} should be larger owing to the hyperfine broadening, i.e., they should deviate from the smooth curves describing the widths of the same lines in U^{238} , Pu^{239} , and Am^{241} . Figure 1 shows the experimental data from^[8]. Its authors reached the conclusion that the splitting responsible for the broadening of the Np^{237} lines is hyperfine splitting and is double the theoretical value²⁾. In our investigation of this discrepancy, we analyzed the experimental data of^[8]. The broadening of each line was defined as the difference between the Np^{237} line width and the width at $Z = 93$ on a parabola drawn through the points for U, Pu, and Am. Then, recognizing that the investigated lines have almost a Lorentz shape, the broadenings were converted into splittings. The weighted average of three values of the splitting of the L_{II} level was 0.32 ± 0.032 Ry. This is not two times but 4.5 times the theoretical estimate of^[8], thus pointing to a recalculation error in this reference. Furthermore, if we use the more exact value $\mu(Np^{237}) = 3.3$ ^[12], then the discrepancy becomes even worse. Finally, calculating the splitting of the L_{II} level with the aid of the more exact formula (4), we obtain a value 0.039 Ry, which leads, even without corrections for the finite dimensions of the nucleus and for screening, to an eight-fold increase of the splitting observed in^[8] over the theoretical value. We note that the broadenings corresponding to a theoretical splitting of 0.039 Ry (they are marked by squares drawn to scale at the corresponding curves in Fig. 1) turn out to be several times smaller than the errors with which the widths of the investigated lines were measured in^[8].

Further analysis of the data of Merill and DuMond led to one more contradiction. If the broadening of the investigated Np^{237} lines is attributed to the action of the HFI effect on the L_{II} level, then the lines corresponding

²⁾The theoretical value was determined in [8] from the formulas for the optical levels with $\mu(Np^{237}) = 6.0$, and turned out to equal 0.07 Ry (1 Ry = 13.6 eV).

to the transition to the L_I level must broaden in the same Np^{237} . The ratio of the HFI splitting of the L_I level to the hyperfine splitting of the L_{II} level, calculated from the formulas in (4), turns out to be 3.3 in the case of Np^{237} . Correction for the finite dimensions of the nucleus lowers this ratio to ≈ 2.7 . The data of^[8] include a dependence of the line width L_{β_4} ($M_{II} \rightarrow L_I$) on Z (it is shown in the upper part of Fig. 1). By determining from the widths of this line for U, Pu, and Am (by linear interpolation) the width of the Np^{237} line not subject to the action of the HFI effect, and by introducing the hyperfine broadening calculated from $\Delta(L_{II})$, we find that the width of the L_{β_4} line should be 2.45 ± 0.01 Ry (point A in Fig. 1)³⁾. This contradicts the experimental value 1.90 ± 0.08 Ry (point B on Fig. 1).

We can thus conclude that the broadening of the L_{II} level of Np^{237} , observed by Merrill and DuMond^[8], is not the consequence of the HFI effect, and that at their accuracy they could not have observed an HFI effect close to the theoretical value. The explanation of the broadening of the L_I level of Np^{237} can be sought among the multielectron effects (cf, e.g.,^[13]).

EXPERIMENT

The experiment was performed with a crystal-diffraction spectrometer in a Cauchois geometry, described in^[14], but in this case one receiving slit was placed in front of the detector, and the line profile was obtained by successive rotations of the crystal through the Bragg angle ϑ_B . For each ϑ_B position, the compared samples, which were rotated in order to average out the possible inhomogeneities^[14], were introduced in succession into the field of view of the instrument. The possible influence of the aftereffect^[15] was cancelled out by varying the sequence of the samples. Two experiments were performed. In the first we investigated the K_{α_1} lines of Eu^{151} and Eu^{153} ^[16], and in the second the K_{α_1} lines of Sb^{121} and Sb^{123} . The widths of the receiving slits were determined from the condition that the measurement time necessary to observe the sought effect at the given accuracy be minimal, and turned out to be 155 and 160 μ for Eu and Sb, respectively. We used the reflection of x-ray quanta from the (1340) plane of quartz. The quasi-mosaic^[17] distance was 160 μ on the focal circle. The apparatus was carefully adjusted before the start of each experiment. The tolerances were calculated in such a way that the possible systematic errors did not exceed several per cent. For example, in the case of europium we have $|\Delta R| \lesssim 2$ mm, where ΔR is the inaccuracy in positioning the receiving slit along the radius (adjustment of the focal distance); $|\Delta\varphi| \lesssim 1'$, where $\Delta\varphi$ is the angle between the receiving slit and the focused line; and $|\Delta h| \lesssim 2.5$ mm, where Δh is the deviation of the height of the slit center from the plane of the focal circle. The relative intensities of the compared samples varied on the average within $\pm 0.5\%$. The line widths at half-height were ~ 40 eV (25") for europium and ~ 17 eV (26") for antimony. The counting rates at the maxima of the lines were $50,000 \text{ min}^{-1}$ at a background of 500 min^{-1} in both experiments. A total of

4×10^8 counts was accumulated for each europium or antimony isotope.

In the first experiment, three europium sources were made of a mixture enriched with Eu^{151} (97.5%), and three enriched with Eu^{153} (99.3%); all six were in the form of the compound Eu_2O_3 . Analogously, in the second experiment we prepared one set of three sources of Sb^{121} (99.6%) and another of Sb^{123} (97.9%), but in all six samples the antimony was in metallic form. To obtain microscopic ingots of Sb^{121} and Sb^{123} , the initial isotope samples, in the form of oxides, were remelted in a melt of Rochelle salt and borax.

The optimal (cf, e.g.,^[14]) weights of Eu_2O_3 and antimony were mixed with polyethylene and pressed into pellets. To prevent a possible influence of different chemical states of samples of the compared isotopes, the chemical state was standardized periodically during the course of each experiment. The accompanying repressing of the pellets led to an averaging of the residual aberration distortions of the line shapes.

The measurement results were mathematically reduced with a computer. From the difference of the experimental curves of the K_{α_1} lines of the compared isotopes we determined simultaneously the values of the isotopic shift δ and the square of the hyperfine splitting Δ^2 , by fitting this difference to the $\tilde{\Delta}F(x)$ curve by least squares (cf. A.3); the quadratic functional was minimized by varying the three parameters $\tilde{\Delta}A$, δ , and Δ^2 . The function $f(x, x_0)$ was used in the form (A.4), in which the parameters x_0 and γ were obtained likewise by least squares in each measurement run prior to reducing the difference curve. Thus, the sought result of each individual measurement run consisted of three independent values of δ and Δ^2 with their errors (in accord with the three compared sample pairs). Further reduction of the experimental data was carried out formally by somewhat different methods for europium and antimony. In the case of europium, the entire sum of the experimental data was broken up into five large series (in the order in which they were obtained). In each large series we obtained the weighted mean values and their errors.

As an additional control over the chemical state of the compared samples, we measured after the line K_{β_1} ($3p_{1/2} \rightarrow 1s_{1/2}$) each larger series. The chemical shift of the K_{β_1} line (due to the possible residual chemical non-identity of the samples made from the compared isotopes) was 2.26 times larger than for K_{α_1} ^[18], and the isotopic shift and the hyperfine broadening were the same, since they were determined practically completely by the $1s_{1/2}$ level. The joint reduction made it possible to obtain separately the values of the effect of the isotopic and chemical shifts δ_{is} and δ_{ch} , so that we were able to monitor the identity of the chemical state of the isotopes during the course of the experiment. The results of the reduction are summarized in the table⁴⁾. Figure 2 shows the resultant positions and widths of the

⁴⁾It should be noted that the final value $\Delta^2 = 1.50 \pm 0.27 \text{ eV}^2$, given in the table was obtained with the aid of formula (A.3) under the assumption that $\mu(Eu^{153}) = 0$. Therefore the final result for the hyperfine splitting of Eu^{151} contains a correction of 10% to take this circumstance into account.

³⁾We recall (see the Appendix) that the line broadening is approximately proportional to the square of the splitting.

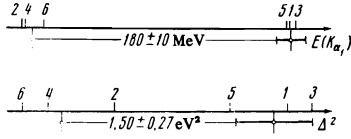


FIG. 2. Positions and squares of the splittings of the $K_{\alpha 1}$ lines from the compared samples. Samples 1, 3, 5—Eu¹⁵¹, 2, 4, 6—Eu¹⁵³.

$K_{\alpha 1}$ lines from individual sources, from which it is possible to judge the degree to which there are no false effects.

In the case of antimony, the results of the individual series were not combined into larger series, and the mean values of δ and Δ^2 and their errors were determined directly from the entire aggregate of the data. We calculated the errors from the mean-squared scatter of the results of individual series relative to the mean value

$$\sigma_{\text{ext}} = \left[\frac{1}{n-1} \sum \frac{(x_i - \bar{x})^2}{\sigma_i^2} / \sum \frac{1}{\sigma_i^2} \right]^{1/2}$$

and the errors of the same weighted mean values

$$\sigma_{\text{int}} = \left(\sum \frac{1}{\sigma_i^2} \right)^{-1/2},$$

where σ_i takes into account only the normal statistical fluctuations of the accumulated counts. As a result, we obtained

$$\delta = 0.3 \pm 1.0(1.0) \text{ meV}, \quad \Delta^2 = 0.45 \pm 0.10(0.09) \text{ eV}^2 \quad (8)$$

The parentheses contain the values of σ_{int} . Figure 3 shows histograms of the experimental values of Δ^2 and δ relative to their mean weighted values. The smooth curves correspond to a Gaussian distribution plotted with respect to the same weighted mean values with a variance $D = \sigma_{\text{int}}^2$. The good agreement between the histograms and the Gaussian distributions shows that there are no random factors capable of shifting the estimated weighted mean values. The values of Δ were calculated under the assumption that $\mu(\text{Eu}^{151})/\mu(\text{Eu}^{153}) = 2.26$, $\mu(\text{Sb}^{121})/\mu(\text{Sb}^{123}) = 1.32$, and that the spins of Eu¹⁵¹, Eu¹⁵³, and Sb¹²¹ are equal to 5/2, and that of Sb¹²³ is 7/2^[12].

The final result of the experiment is

$$\Delta(\text{Eu}^{151}) = 1.36 \begin{matrix} +0.11 \\ -0.13 \end{matrix} \text{ eV}, \quad \Delta(\text{Sb}^{121}) = 0.68 \begin{matrix} +0.07 \\ -0.08 \end{matrix} \text{ eV}, \quad (9)$$

$$\delta(\text{Eu}^{151-153}) = 186 \pm 10 \text{ meV}, \quad \delta(\text{Sb}^{121-123}) = 1.8 \pm 1.0 \text{ meV}. \quad (10)$$

The values of Δ and their errors correspond to the mean values of Δ^2 and their variances. The results (9)

Results of individual (large) measurement series for Eu¹⁵¹⁻¹⁵³

No. Series	δ_{ch} , meV	δ_{is} , meV	Δ^2 , eV ²
1	-8 ± 23	191 ± 21	1.57 ± 0.47
2	-6 ± 27	191 ± 28	1.35 ± 0.58
3	22 ± 13	150 ± 17	1.87 ± 0.61
4	-41 ± 38	218 ± 39	1.13 ± 0.94
5	7 ± 19	195 ± 19	1.19 ± 0.83
Average	8 ± 9	180 ± 10	1.50 ± 0.27

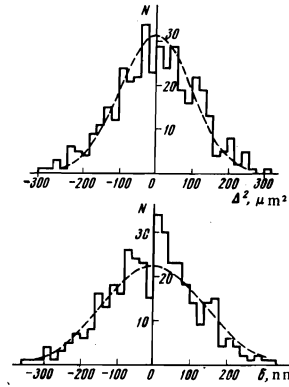


FIG. 3. Histograms of the experimental values relative to the corresponding mean weighted values in the case of antimony. N—number of events in the given channel.

and (10) take into account the isotopic composition of the investigated samples and the isotopic mass shift^[22].

The theoretical values of the splittings were obtained from formulas (3) and (6). For $\mu(\text{Eu}^{151}) = 3.464$ and $\mu(\text{Sb}^{121}) = 3.359$ ^[12] we get

$$\Delta_{\text{theor}}(\text{Eu}^{151}) = 1.46 \text{ eV}, \quad \Delta_{\text{theor}}(\text{Sb}^{121}) = 0.67 \text{ eV}. \quad (11)$$

As seen from a comparison of (9) and (11), the theoretical data agree well with experiment.

ISOTOPIC SHIFTS

Let us examine the values of the isotopic shifts of the $K_{\alpha 1}$ lines between the isotopes Eu¹⁵¹, Eu¹⁵³ and Sb¹²¹, Sb¹²³ obtained in the present investigation (cf. (10)). These shifts are due to the changes in the mean-squared charge radius of the nuclei following the addition of two neutrons to one of the compared isotopes. Usually the shifts yield directly the changes of the mean-squared radii themselves, in relative units:

$$\gamma = \Delta \langle R^2 \rangle / \Delta \langle R^2 \rangle_{R \sim A^{1/3}}.$$

It is convenient to use the quantity γ in comparisons of $\Delta \langle R^2 \rangle$ obtained by different experimental and theoretical methods.

Using (10) and Babushkin's data^[19], we obtain

$$\gamma(\text{Eu}^{151-153}) = 2.87 \pm 0.15, \quad \gamma(\text{Sb}^{121-123}) = 0.08 \pm 0.05. \quad (12)$$

The value of γ for the Eu¹⁵¹—Eu¹⁵³ pair, being anomalously large (the largest of all the known value^[1]), could previously be obtained only from data on the isotopic shift of optical lines^[20], where there was the danger of an influence of non-nuclear multielectron effect^[1]. It was equal to 2.98 ± 0.32 . A comparison of this value with (12) shows that the optical and x-ray data on the isotopic shifts are in good agreement.

The case of antimony is of interest because, according to the theoretical data of Bunatyan^[21], who used the theory of finite Fermi systems, an anomalously small change of the charge radius should be observed on going from Sb¹²¹ to Sb¹²³ (the number of neutrons changes from 70 to 72) (the value of γ should be 0.22, whereas for the neighboring elements (tin and tellurium), with the same change in the number of neutrons, γ turns out to be ~ 0.48 ^[22]). This anomaly is attributed to the fact that the addition of two neutrons in the antimony nuclei

is accompanied simultaneously by a changeover of the odd proton from the state $2d_{5/2}$ (in Sb^{121}) to the state $1g_{7/2}$ (in Sb^{123})^[23]. The appreciable increase of the orbital momentum of the odd proton (from 2 to 4) decreases the polarization of the proton core by the odd proton, and this compensates to a considerable degree for the increase of the charge radius by the addition of the neutron pair. Our experimental value of γ (see (12)) turns out to be even somewhat smaller than the theoretically expected one. Thus, the allowance in^[21] for the change in the concrete proton states describes correctly the character of the redistribution of the charge density in the nucleus.

The discussed value of γ can be obtained also from data on the optical isotopic shift^[24]. In this case it turns out that $\gamma_{\text{opt}}(\text{Sb}^{121-123}) = 0.05$. The possible presence of a specific mass shift^[1], generally speaking, does not make it possible to estimate the change of the mean-squared charge radius of the nucleus from $\gamma_{\text{opt}}(\text{Sb}^{121-123})$ with sufficient reliability. In this case, however, γ_{opt} agrees well with the value in (10). The specific mass shift, which can be easily obtained from a comparison of the optical data in our data, is small and lies in the interval $0-0.02 \text{ cm}^{-1}$.

CONCLUSION

Insofar as we know, we have experimentally observed for the first time the HFI effect of broadening of spectral lines. The experimentally determined splittings are in good agreement with the theoretical ones.

We have obtained at the same time data on the changes of the charge distributions in the nuclei on going from Sb^{121} to Sb^{123} and from Eu^{151} to Eu^{153} . In the case of antimony, the experiment concerns the anomaly predicted for $\text{Sb}^{121-123}$ within the framework of the theory of finite Fermi systems. In the case of europium, the validity of the previously obtained optical data was confirmed.

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APPENDIX

The shape of the apparatus curve of a single line can be written in the form

$$F(x, x_0) = A_0 f(x, x_0), \quad (\text{A.1})$$

where A_0 is the amplitude and is proportional to the transmission, x is the running coordinate, and x_0 is the position of the center of gravity of the line. The line shape of the hyperfine-structure doublet (the hyperfine splitting occurs without a shift of the center of gravity), shifted by an amount δ relative to the point x_0 , then takes the form

$$F(x, x_0, \delta, \Delta, C) = A \left[\frac{1}{1+C} f\left(x, x_0 + \delta - \frac{\Delta C}{1+C}\right) + \frac{C}{1+C} f\left(x, x_0 + \delta + \frac{\Delta}{1+C}\right) \right],$$

where C is the ratio of the intensities of the components of the doublet ($C = I/(I+1)$), Δ is the magnitude of the hyperfine splitting. Assuming that δ and Δ are small in comparison with the line width and that $\delta^2 \ll C\Delta^2/(1+C)^2$, we have

$$F(x, x_0, \delta, \Delta, C) = A \left[f(x, x_0) + \delta f'(x, x_0) + \frac{C}{2(1+C)^2} \Delta^2 f''(x, x_0) \right], \quad (\text{A.2})$$

where f' and f'' are the first and second derivatives with respect to x_0 .

It is seen from (A.2) that the deformation of the profile, due to the splitting, takes the form of line broadening, since $f''(x, x_0)$ is maximal and negative in the region $x \approx x_0$, and is small in the region corresponding to the half-width of the $f''(x, x_0)$ curve. The broadening is proportional to the square of the splitting Δ .

For the difference $\tilde{\Delta}F(x)$ we can write, if both compared lines are HFI doublets,

$$\tilde{\Delta}F(x) = F(x, x_0, \delta, \Delta, C) - F(x, x_0, \Delta_0, C_0) = f(x, x_0) \tilde{\Delta}A + A \delta f'(x, x_0) + \frac{A}{2} \left[\frac{C}{(1+C)^2} - \frac{C_0}{(1+C_0)^2} \frac{\Delta_0^2}{\Delta^2} \right] \Delta^2 f''(x, x_0). \quad (\text{A.3})$$

Here $\Delta A = A - A_0$ and the ratio Δ_0^2/Δ^2 is proportional to the ratio μ_0^2/μ^2 of the squares of the magnetic moments of the nuclei of the compared isotopes. In the present paper, the function $f(x, x_0)$ of formula (A.1) was of the form

$$f(x, x_0) = (z+a) \text{arctg}(z+a) + (z-a) \text{arctg}(z-a) - (z+b) \text{arctg}(z+b) - (z-b) \text{arctg}(z-b) + \frac{1}{2} \ln \frac{[1+(z+b)^2][1+(z-b)^2]}{[1+(z+a)^2][1+(z-a)^2]}, \quad (\text{A.4})$$

where $z = 2(x - x_0)/\gamma$, $a = (t+s)/\gamma$, $b = (t-s)/\gamma$. Expression (A.4) can be obtained by successive integration of the Lorentz profile of an x-ray line with natural width γ , a quasisosaic structure^[17] of the reflecting crystal with width t much larger than the natural mosaic, and an instrument receiving-slit width s ; this accounts for the principal part of the apparatus line shape of the x-ray line in the crystal-diffraction instrument. The possible corrections may be due to the inaccuracy of the focusing of the instrument, to aperture aberrations, and other causes of profile deformation. If the apparatus is suitably adjusted, these corrections can be made sufficiently small (see the section entitled "Experiment"). It should be noted that in the case of the method in which the sources are alternately introduced into the field of view of the instrument^[14] used in the present study, the influence of such deformations of the profile is particularly small, since they are practically the same for the two compared lines, and the corresponding difference of the counting rates is described by the curve (A.3).

¹H. Kopfermann, Nuclear Moments, Academic, 1958.

²R. J. Powers, Phys. Rev. **169**, 1 (1968).

³G. Breit, *ibid.* **35**, 1447 (1930).

⁴F. Richtmeyer and S. Barnes, *ibid.* **37**, 1965 (1931).

⁵J. Williams, *ibid.* **37**, 1431 (1931).

⁶M. Frillay, B. Gohkale and M. Voladares, CR, Paris, **283**, 1183 (1951).

⁷G. Rogosa and G. Schwarz, Phys. Rev. **92**, 1434 (1953).

- ⁸ J. Merrill and J. DuMond, *Ann. Phys.* **14**, 166 (1961).
- ⁹ F. A. Babushkin, *Zh. Eksp. Teor. Fiz.* **42**, 1604 (1962) [*Sov. Phys.-JETP* **15**, 1113 (1962)].
- ¹⁰ M. Wilson, *Phys. Rev.* **176**, 58 (1968).
- ¹¹ J. C. Slater, *ibid.* **36**, 57 (1930).
- ¹² G. H. Fuller and V. W. Cohen, *Nucl. Data A5*, 433 (1969).
- ¹³ V. I. Nefedov, *Zhurnal strukturnoi khimii* **7**, 4 (1966).
- ¹⁴ O. I. Sumbaev, A. F. Mezentsev, V. I. Marushenko, A. S. Ryl'nikov and G. A. Ivanov, *Yad. Fiz.* **9**, 906 (1969) [*Sov. J. Nucl. Phys.* **9**, 529 (1969)].
- ¹⁵ O. I. Sumbaev and A. F. Mezentsev, *Zh. Eksp. Teor. Fiz.* **49**, 459 (1965) [*Sov. Phys.-JETP* **22**, 323 (1966)].
- ¹⁶ A. S. Ryl'nikov, G. A. Ivanov, V. I. Marushenko, A. I. Smirnov and O. I. Sumbaev, *ZhETF Pis. Red.* **12**, 128 (1970) [*JETP Lett.* **12**, 88 (1970)].
- ¹⁷ O. I. Sumbaev, *Zh. Eksp. Teor. Fiz.* **54**, 1352 (1968) [*Sov. Phys.-JETP* **27**, 724 (1968)].
- ¹⁸ O. I. Sumbaev, Yu. P. Smirnov, E. V. Petrovich, V. S. Zykov and A. I. Grushko, *ZhETF Pis. Red.* **10**, 209 (1969) [*JETP Lett.* **10**, 131 (1969)].
- ¹⁹ F. A. Babushkin, *Optika i spektroskopiya* **15**, 721 (1963).
- ²⁰ P. Brix and H. Kopferman, *Rev. Mod. Phys.* **30**, 517 (1958).
- ²¹ G. G. Bunatyan, *Yad. Fiz.* **4**, 707 (1966) [*Sov. J. Nucl. Phys.* **4**, 502 (1967)].
- ²² S. K. Bhattacharjee, F. Boehm and P. L. Lee, *Phys. Rev.* **188**, 1919 (1969).
- ²³ Translation in: *Stroenie atomnogo yadra (Structure of Atomic Nuclei)*, III, 1959.
- ²⁴ D. H. Tomboulion and R. F. Bacher, *Phys. Rev.* **58**, 52 (1940).

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