

time  $\sim \Gamma^{-1} \sim 10^{-7}$  sec all the atoms are again in the ground state and, therefore, the distribution of the atomic velocities at the lower active level no longer has a Bennett dip (hole) associated with the resonant nature of the excitation of atoms. In spite of this, for an additional time of  $\sim \gamma^{-1} \sim 10^{-1}$  sec the momentum distribution still has a dip of half-width (24), which is explained by a redistribution of the velocities of atoms under the influence of light. This can be discovered from the resonant reduction in the absorption by a test light wave in the case of exact tuning to one of the allowed transitions from the ground state. An experiment of this kind can also be used to resolve closely spaced spectral lines masked by the Doppler broadening. Moreover, there is a possibility of observing directly the process of relaxation of the nonequilibrium distribution of the atomic velocities to its equilibrium form, i.e., in the final analysis this makes it possible to study collisions of atoms in a gas (for example, the reciprocal of the time needed for the disappearance of a dip is equal to the frequency of the velocity-changing atomic collisions). Information on atomic collisions is contained also in the steady-state dip profile.

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## Hyperfine shift of x-ray lines excited in internal conversion

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The energy shift of x-ray lines following internal conversion, which is due to the nonstatistical population of the hyperfine structure sublevels, is discussed. The nonstatistical population arises from the interaction of the magnetic moment of the nucleus in the final state with the electron current in the atomic shell ( $K$  or  $L$ ) that has a single vacancy. Expressions are obtained for the corresponding populations and shifts for the case of conversion transitions of arbitrary multipolarity. It is pointed out that from the experimental values of the shift one can derive the magnetic moment of the nucleus (provided the conversion-transition parameters are known) or the characteristics of the conversion transition itself (provided the nuclear magnetic moment is known). The shift of the barium  $K_{\alpha 1}$  line has been measured on isotopically enriched specimens, and the previously unknown magnetic moment of the excited state of the  $^{133}\text{Ba}$  nucleus has been determined from the results as  $+0.51 \pm 0.07$  nuclear magneton.

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The effect of the nonstatistical population of hyperfine-structure (HFS) sublevels of x-ray terms of atoms excited as a result of internal conversion was first noted in Ref. 1, where it was used to determine the magnetic

moment of a nucleus.

In the first part of the present paper we present a theoretical description of the effect and discuss its influ-

ence on the energy shifts of the x-ray emission lines<sup>1)</sup> that follow the internal conversion process. In the second part of the paper we describe an experiment to determine the magnetic moment of an excited state of the <sup>133</sup>Ba nucleus, using known crystal-diffraction techniques<sup>2</sup> for measuring small energy shifts of x-ray lines.

## THEORY

We shall calculate the probability  $W_e^F$  for excitation of a definite HFS sublevel ( $F$ ) of the final atom resulting from internal conversion. Let us consider the following process: the atom undergoes a transition from an initial state with total angular momentum  $I_0$  (the nucleus is in an excited state with spin  $I_0$  and the electron shell (the  $K$  and  $L$  electrons) is closed with zero angular momentum) to a final state (in which the nucleus has spin  $I$  and the corresponding shell has one vacancy and angular momentum  $j_0$ , the total angular momentum of the atom being<sup>2)</sup>  $F=I+j_0$ ) with emission of an electron into the continuum with angular momentum  $j$ . The matrix element for such a transition can be written in the form

$$H_e^F = \sum_{M, \mu, m_0, m} C_{j_0 m_0 M}^{I_0 0} C_{j_0 m_0 \mu}^{F M} \langle \Psi_{I_0 \mu} \Psi_{j_0 m_0} | H_e | \Psi_{I_0 \mu} \Psi_{j_0 m_0} \rangle (-1)^{j_0 + m_0} \\ = \sum_{M, \mu, m_0, m} C_{j_0 m_0 M}^{I_0 0} C_{j_0 m_0 \mu}^{F M} \langle \Psi_{I_0 \mu} \Psi_{j_0 m_0} | H_e | \Psi_{I_0 \mu} \Psi_{j_0 m_0} \rangle (-1)^{j_0 - m_0}, \quad (1)$$

where  $m$ ,  $m_0$ ,  $\mu$ ,  $\mu_0$ , and  $M$  are the projections of the angular momenta  $j$ ,  $j_0$ ,  $I$ ,  $I_0$ , and  $F$ , respectively;  $C_{j_1 m_1 j_2 m_2}^{j_3 m_3}$  is a Clebsch-Gordan coefficient;  $\Psi_{I_0 \mu}$  and  $\Psi_{I \mu}$  are the initial- and final-state wave functions of the nucleus while  $\Psi_{j_0 m_0}$  and  $\Psi_{j m}$  are those of the electron; and  $H_e$  is the operator for the conversion transition. The matrix element on the right in Eq. (1) is well known from the theory of internal conversion (see, e.g., Ref. 3, pp. 25 and 34) and has the form

$$\langle \Psi_{I_0 \mu} \Psi_{j_0 m_0} | H_e | \Psi_{I_0 \mu} \Psi_{j_0 m_0} \rangle = \sum_{L \Lambda} C_{I_0 \mu \Lambda L}^{I_0 0} C_{j_0 m_0 \Lambda}^{j_0 m_0} b_{\kappa}(\tau L). \quad (2)$$

Here  $L$  and  $\Lambda$  are the transition angular momentum (multipolarity) and its projection, respectively ( $|I_0 - I| \leq L \leq I_0 + I$ );  $\tau$  specifies the type of the transition ( $\tau = M$  for a magnetic transition and  $\tau = E$  for an electric transition); and  $\kappa$  characterizes the final state of the ejected (continuum) electron in the central field:  $\kappa = (l + j)(2j + 1)$  and  $j = |\kappa| - (\frac{1}{2})$ , where  $l$  and  $j$  are the orbital and total angular momenta of the electron, respectively.

On substituting (2) into (1) and performing the summation over the magnetic quantum numbers, we obtain (except for an unimportant phase factor) the matrix element for the conversion transition to a definite HFS state ( $F$ ) of the final atom:

$$H_e^F = [(2F+1)(2j+1)]^{1/2} \sum_L (-1)^L \left\{ \begin{matrix} I_0 & I & L \\ j_0 & F & L \end{matrix} \right\} b_{\kappa}(\tau L), \quad (3)$$

where the curly brackets denote the Wigner  $6j$  symbol.<sup>4</sup> The desired probability  $W_e^F$  itself is given by

$$W_e^F = \frac{2\pi}{\hbar} \sum_{\kappa} |H_e^F|^2. \quad (4)$$

The shift of the center of gravity of an x-ray line accompanying conversion with respect to that of a line emitted in the absence of hyperfine interaction is

$$\Delta E = \sum_F \Delta^F W_e^F / \sum_F W_e^F. \quad (5)$$

where  $\Delta^F$  is the hyperfine shift of the sublevel  $F$ . Formula (5) is valid if the hyperfine splitting of the final term of the x-ray transition can be neglected. We note that  $\Delta E$  vanishes in the case of a statistical population (when the probability  $W_e^F$  is proportional to the statistical weight  $2F+1$  of the final state of the atom). Such a situation arises, for example, in the photoexcitation of an atom,<sup>5</sup> and this makes it possible to use fluorescence lines as reference standards in experiments. In this case  $\Delta E = E^{\text{conv}} - E^{\text{phot}}$ , where  $E^{\text{conv}}$  and  $E^{\text{phot}}$  are the energies of the x-ray lines excited in internal conversion and by photoexcitation, respectively.

As a specific example we shall derive the expression for the shift  $\Delta E^{K\alpha_1}$  of the  $K\alpha_1$  line (arising from the transition between the  $K$  and  $L_{III}$  x-ray terms) excited as a result of an internal conversion transition of specified multipolarity  $\tau L$ . Then  $j_0 = \frac{1}{2}$ ,  $\Delta^{F=I-1/2} = -\Delta_K(I+1)/(2I+1)$ , and  $\Delta^{F=I+1/2} = \Delta_K I/(2I+1)$ ,<sup>5,6</sup> where  $\Delta_K$  is the hyperfine splitting of the  $K$  level<sup>3)</sup>:

$$\Delta_K = \alpha E_0 \left( \frac{m_p}{m_e} \right) \mu_I \frac{2I+1}{I} \frac{2(\alpha Z)^3}{3\gamma(2\gamma-1)} (1 - e_e - e_m), \quad (6)$$

in which  $\alpha$  is the fine structure constant,  $Z$  is the charge number of the nucleus,  $[\gamma = 1 - (\alpha Z)^2]^{1/2}$ ,  $m_e/m_p$  is the electron: proton mass ratio, and  $E_0 = m_e c^2$  is the electron rest energy,  $\mu_I$  is the magnetic moment of the nucleus in nuclear magnetons, and  $\omega_e$  and  $\omega_m$  are corrections for the distributions of charge and magnetization within the nucleus. Equation (6) is accurate to within 1%. Using Eqs. (3)–(5) in this case, we obtain

$$\Delta E^{K\alpha_1} = \Delta_K \frac{(I_0 - I)(I_0 + I + 1) - L(L + 1)}{2L(2I + 1)} \frac{1 - \rho}{1 + (L + 1)\rho/L}, \quad (7)$$

where  $\rho = |b_{\kappa_2}(\tau L)|^2 / |b_{\kappa_1}(\tau L)|^2$ ,  $|\kappa_1| < |\kappa_2|$ , and  $L > 0$  ( $\kappa_1$  and  $\kappa_2$  specify the two final continuum states possible for the electron in  $K$  conversion).

In the case of allowed nuclear transitions,  $b_{\kappa}$  is actually determined by matrix elements that are proportional to the amplitude for  $\gamma$ -ray emission and can be calculated fairly well numerically; use can be made of this circumstance to derive the magnetic moment from the hyperfine shift of the x-ray line.

We note that the above method of determining nuclear magnetic moments  $\mu$  differs from the known methods that make use of external magnetic fields in that the magnetic field acting on the nucleus can be calculated accurately enough since it is due to the current of the inner atomic electrons. Thus, the proposed method makes it possible to measure  $\mu$  directly. This is also of interest in connection with the experimental study of the physics of atomic effects that may, for example, strengthen or weaken external magnetic fields in the vicinity of the nucleus. In the case of strongly hindered transitions for which the amplitude for  $\gamma$ -ray emission is small (the case of anomalous conversion) the main contribution to  $b_{\kappa}$  comes from intranuclear conversion matrix elements, which are calculated on the basis of definite nuclear-structure models. In this case, if the magnetic moment of the final state of the nucleus is

known the line shift provides information on the adequacy of the model used in the calculation, which supplements the information that can be obtained by known methods from conversion-coefficient measurements and correlation experiments.

We also note that in the case of a mixed transition the expression for  $W_s^{\mathcal{P}}$  contains an interference term due to interference between transitions of different multiplicities (there is no such term in the total conversion coefficients). The expression for the shift  $\Delta E$  therefore contains a term that is linear in the multipole mixing parameter  $\delta_\gamma$ .<sup>3</sup> This may prove to be useful in studying mixed transitions.<sup>4)</sup>

For ordinary (not anomalous) conversion, expression (7) for the shift reduces (when using the explicit form of the matrix elements  $b_\kappa$ , see p. 25 of Ref. 3) to the form

$$\Delta E^{K\alpha_1} = \Delta_\kappa \frac{(I_0 - I)(I_0 + I + 1) - L(L + 1) \frac{1 - Lr/(L + 1)}{1 + r}}{2L(2I + 1)}, \quad (8)$$

where  $r = |M_{\kappa_2}(\tau L)|^2 / |M_{\kappa_1}(\tau L)|^2$ ,  $M_\kappa(\tau L)$  being the partial conversion matrix element [ $\alpha(\tau L) = \sum_\kappa |M_\kappa(\tau L)|^2$ , where  $\alpha(\tau L)$  is the total coefficient for conversion on a definite atomic shell or subshell<sup>3,8)</sup>]. The  $M_\kappa(\tau L)$  have been partially tabulated,<sup>8</sup> or they may be calculated with special computer programs (see, e.g., Ref. 9). Equation (8), together with Eq. (6), yields an equation for the magnetic moment of the nucleus in the final state. In the case of mixed transitions, the expressions for the shifts are more cumbersome, but they can always be obtained from Eqs. (3) and (4) when the multipole mixing parameter  $\delta_\gamma$  is known. We note that the shifts given in Ref. 1 for M1 transitions correspond to Eq. (8) with  $r = 0$ . This approximation is accurate within 10% for all ML transitions with energies up to 0.5 MeV in nuclei with  $Z \geq 55$ .

## EXPERIMENT

To determine the unknown magnetic moment of the 12.3 keV  $3/2^+$  excited state  $^{133}\text{Ba}^*$  of the  $^{133}\text{Ba}$  nucleus, whose lifetime<sup>10</sup> is  $\tau = 6.8 \pm 0.4$  nsec, we measured the energy shift of the barium  $K_{\alpha_1}$  line accompanying internal conversion of the 276 keV M4 nuclear transition  $^{133}\text{Ba}^m(11/2^- \rightarrow 3/2^+)^{133}\text{Ba}^*$ . The  $^{133}\text{Ba}^m$  isomer was obtained by exposing a  $\text{BaCO}_3$  specimen enriched in  $^{132}\text{Ba}$  (the isotopic compositions of the specimens used in the study are given in Table I) to a  $\sim 10^{14}$   $\text{cm}^{-2}\text{sec}^{-1}$  neutron flux at the reactor. The barium  $K_{\alpha_1}$  fluorescence line excited by radioactive  $^{170}\text{Tm}$  in a  $\text{BaCO}_3$  specimen enriched in  $^{135}\text{Ba}$  was used as a reference standard. The measurements were made with a Cauchois crystal-diffraction spectrometer according to the scheme that we ordinarily employ (see, e.g., Ref. 2).

A new setup was constructed, which differed from

earlier setups in that the source exchanger was suitable for work with activities up to 100 Ci. To increase the luminosity of the spectrometer, the height of the entrance slit to the detector was doubled and two FEU-93 photomultipliers with NaI(Tl) crystals 45 mm in diameter were mounted behind the slit, one above the other and symmetrically disposed with respect to the plane of the focal circle. For the same purpose (i.e., to minimize the time required to measure the shifts of the barium  $K_{\alpha_1}$  line) we optimized the cut parameters of the single-crystal quartz plate, so that reflection was from the (203) planes, which were perpendicular to the large faces and parallel to the small faces of the plate. The bending coefficient  $k$  of the reflecting planes<sup>2</sup> turned out to be  $k = 2 \times 10^{-4}$   $\text{cm}^{-1}$ ; the plate was 1.2 mm thick, the entrance slit to the detector was 0.4 mm wide, and the focal circle was 2 m in diameter. As a result of all this, the spectrometer luminosity increased by a factor of three [as compared with the use of a single detector and a plate cut in the standard way with reflection from the (130) planes<sup>2</sup>].

The specimens to be compared consisted of wafers 20 mm in diameter and 2–3 mm thick and were introduced alternately into the field of view of the spectrometer. Statistics were accumulated for a total of  $\sim 120$  hr in separate runs of 1–2 hr each. Because of the short half-life of the  $^{133}\text{Ba}^m$  isomer ( $T_{1/2} = 38.9$  hr) the shape of the barium  $K_{\alpha_1}$  line changed during a single run, so a correction for the radioactive decay of  $^{133}\text{Ba}^m$  was included in the data processing procedure. The experimental shift turned out to be  $+57.8 \pm 7.4$  (6.8) meV (two errors are shown, the external and internal rms deviations, respectively).

The principal difficulty inherent in the present method is that of distinguishing the  $K_{\alpha_1}$  radiation corresponding to the investigated conversion transition against the background of such other barium  $K_{\alpha_1}$  radiation as may present. In the case of  $^{133}\text{Ba}^m$ , all the  $K_{\alpha_1}$  radiation emitted following its decay corresponds to the investigated 276M4 transition. The only source of extraneous barium  $K_{\alpha_1}$  radiation is the  $^{134}\text{Ba}$  present as an impurity in the  $^{132}\text{Ba}$  specimen. The isomer  $^{135}\text{Ba}^m$  resulting from the reaction  $^{134}\text{Ba}(n, \gamma)^{135}\text{Ba}^m$  decays to the ground state by the well converted 268M4 ( $11/2^- \rightarrow 3/2^+$ ) transition with the half life  $T_{1/2} = 28.7$  hr. The resulting extraneous barium  $K_{\alpha_1}$  line is also shifted with respect to the fluorescence line as a result of nonstatistical population. This shift can be calculated with formula (8), using the known<sup>11</sup> magnetic moment of the  $^{135}\text{Ba}$  ground state. The admixture of the barium  $K_{\alpha_1}$  line from the  $^{135}\text{Ba}^m$  isomer can also be calculated quantitatively provided the reaction cross section ratio

$$\sigma(^{134}\text{Ba}(n, \gamma)^{135}\text{Ba}^m) / \sigma(^{132}\text{Ba}(n, \gamma)^{133}\text{Ba}^m)$$

and the yields of  $K_{\alpha_1}$  radiation on conversion are known. Unfortunately, the data in the literature on these reaction cross sections are very contradictory,<sup>12,13</sup> so we determined the relative admixture of the barium  $K_{\alpha_1}$  line from the  $^{135}\text{Ba}^m$  isomer experimentally. For this purpose we irradiated  $\text{BaCO}_3$  specimens, enriched in  $^{132}\text{Ba}$  and  $^{134}\text{Ba}$  but otherwise identical, in the reactor under identical conditions and then measured the inten-

TABLE I.

Principal isotope	Composition, %						
	130	132	134	135	136	137	138
132	0.1	28	7.58	8.85	7.28	8.12	40.07
134	<0.06	0.06	85.5	5.98	1.42	1.36	5.47
135	<0.05	<0.05	0.26	92.7	3.62	0.80	2.62

sity ratio of the barium  $K_{\alpha_1}$  lines of the two specimens at the crystal-diffraction spectrometer. Knowing the isotopic compositions of the compared specimens and the half lives of  $^{133}\text{Ba}^m$  and  $^{135}\text{Ba}^m$ , we can calculate the relative contribution to the barium  $K_{\alpha_1}$  line from the  $^{135}\text{Ba}$  isomer in the working specimen; it varied from 4.5 to 2.0% during the entire time in which statistics were being collected. The experimental shift was corrected for the shift of this impurity line; the corrected shift turned out to be  $+57.2 \pm 7.6$  (6.8) meV.

The experimental ratio of the intensities of the barium  $K_{\alpha_1}$  line from the  $\text{BaCO}_3$  specimens enriched in  $^{134}\text{Ba}$  and  $^{132}\text{Ba}$  and irradiated in the reactor was also used to determine the reaction cross section ratio  $\sigma[^{134}\text{Ba}(n, \gamma)^{135}\text{Ba}^m] / \sigma[^{132}\text{Ba}(n, \gamma)^{133}\text{Ba}^m]$ , using the known yields of  $K_{\alpha_1}$  radiation for the 276M4 and 268M4 conversion transitions.<sup>14</sup> This ratio was found to be  $0.18 \pm 0.01$ , which agrees with the value  $0.23 \pm 0.04$  given in Ref. 13, but is in conflict with the value given in Ref. 12.

The measured shift depends on the isotopic and chemical compositions of the specimens (the "isotopic" and "chemical" shifts), as well as on the difference of the volume isotopic shifts between the excited and ground states of the investigated isotope (the "isomeric" shift), and corrections for these effects should be made.

Experimental data from Ref. 15 were used to correct for the isotopic shift; the corrected experimental shift turned out to be  $+55.9 \pm 7.6$  (6.8) meV. The chemical shift was assumed to be zero, since all the specimens had the same chemical form:  $\text{BaCO}_3$ . The isomeric shift can be estimated from the known<sup>16</sup> experimental values of  $\delta\langle r^2 \rangle$  (the increment of the rms charge radius of the nucleus) for the  $3/2^+$  excited states of the neighboring even-odd nuclei  $^{119}\text{Sn}$ ,  $^{125}\text{Te}$ , and  $^{129}\text{Xe}$ . The isomeric shifts of the  $K_{\alpha_1}$  lines of these nuclei corresponding to the experimental  $\delta\langle r^2 \rangle$  values do not exceed 0.3 meV, so no correction was made for the isomeric shift.

On substituting the experimental value of the shift into Eq. (8) we obtain the value  $+0.51 \pm 0.07$  (0.06)  $\mu N$  for the magnetic moment of the  $3/2^+$  state of  $^{133}\text{Ba}^*$ . This value agrees well with the calculated value  $\mu^{\text{calc}} = +0.54$  given by Kisslinger and Sorensen.<sup>17</sup>

In concluding, we note that the possibility of using coincidence techniques or of investigating the shifts of the conversion-electron lines may be of interest in connection with the difficulties discussed above in separating out the x-radiation corresponding to the conversion transition under investigation.

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- <sup>1</sup>We recall<sup>1</sup> that the conversion-electron lines themselves, as well as the corresponding Auger-electron lines, suffer analogous shifts.
- <sup>2</sup>It is not necessary to take the outer shells of the atom into account when  $K$  or  $L$  terms are excited.
- <sup>3</sup>In Ref. 5 the authors made an error in editing: in the expression for the constant  $G$  from formulas (2)–(4) of Ref. 5 the quantity  $\mu_N$  should be replaced by  $m_e/m_p$  (the electron:proton mass ratio). In addition, the small correction for shielding  $\eta$  from Eq. (6) vanishes when it is correctly defined.
- <sup>4</sup>In Ref. 7 it was suggested that the analogous interference between Fermi and Gamow-Teller transitions in the case of  $K$  capture with  $\Delta I = 0$  be used to determine the ratio of the corresponding matrix elements.
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