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COULOMB FRAGMENTATION OF MOLECULES FOLLOWING BETA EMISSION*

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Nuclear resonance-fluorescence scattering from 364-, 637-, and 723-keV levels of Xe^{131} has been observed. The results obtained with a gaseous source of I^{131} were different from those obtained using a gaseous source of CH_3I^{131}. The significant differences are ascribed to the effects of Coulomb fragmentation of the $(IXe^{131})^+$ molecular ion formed following the beta decay of I^{131}.

The enhancement of resonant scattering of gamma rays produced by Coulomb fragmentation has been observed previously in electron-capture decay processes.[1,2] Since beta decay causes much less ionization in the daughter atom than is the case for electron capture, one might well expect Coulomb fragmentation following beta decay to be relatively unimportant compared with that following electron capture.[1,3,4] However, the identification of reaction products following the decay of I^{130} and I^{131} in gaseous methyl and ethyl iodide[5,6] indicates that considerable disruption of the molecules takes place, although the collection times (10^{-4}-10^{-5} sec) involved in these experiments were much longer than the nuclear lifetimes of interest in resonant scattering. In an experiment where resonant scattering from three levels of Xe^{131} (364, 637, and 723 keV) was observed we have found evidence that Coulomb fragmentation plays an important role following the beta decay of I^{131} in gaseous I_2.

In nuclear resonance-fluorescence experiments for the case where the source and absorber nuclei are initially stationary, the emitted gamma ray would be off resonance by an amount $2R_a$, where R_a is the recoil energy imparted to the source and absorber (or scatterer) nuclei. Resonance can be re-established if an initial momentum P_i is imparted to the source nucleus. The emitted gamma-ray energy E_γ is then

$$E_\gamma = E_L - R_a + \vec{P}_i \cdot \vec{P}_\gamma / M_S \rightarrow E_L + R_a \quad \text{for resonance.} \qquad (1)$$

P_γ is the recoil momentum imparted by the gamma ray, E_L, is the energy of the excited level, $R_a = E_\gamma^2/2M_ac^2$, and M_a is the mass of the radioactive-source atom and also the mass of the absorber atom. M_S is the mass of the source molecule. The arrow should be read as "must be made equal to." Equation (1) implies that for resonant scattering to be possible, $P_i/P_\gamma \geq M_S/M_a$.

For the beta decay of I^{131} to the three levels of interest in Xe^{131}, Table I shows the maximum recoil momentum following beta emission, the recoil momentum following gamma emission, the ratio of these momenta, and the ratio of M_S to M_a with sources of HI^{131}, CH_3I^{131}, and II^{131}. Even allowing for the effects of thermal broadening of the emission and absorption lines as well as possible molecular breakup produced by the recoil from the beta emission, it can readily be seen that little if any resonant scattering from

Table I. Recoil momenta and other quantities of interest defined in the text for different molecular sources of I^{131}.

Decay (keV)	P_γ/mc	$P_{i\,\max}/mc$	P_i/P_γ	HI^{131} M_s/M_a	CH_3I^{131} M_s/M_a	II^{131} M_s/M_a
364	0.71	1.93	2.72	1.01	1.11	1.97
637	1.25	1.31	1.05	1.01	1.11	1.97
723	1.41	1.10	0.78	1.01	1.11	1.97

the 723-keV level would be expected.

When the experiment was carried out with a gaseous source of CH_3I^{131}, resonant scattering was observed from both the 364- and 637-keV levels and there was some indication that very weak scattering might have occurred from the 723-keV level as well. On the other hand, with a source in the form of gaseous I_2, the resonant scattering from the 364-keV level was reduced by about a factor of 3 and the scattering from the 637-keV level approximately doubled. There was also the definite appearance of a resonant-scattering peak at 723-keV about half as intense as the 637-keV resonance peak.

Since the experimental results for the I_2 source are clearly inconsistent with those expected under the assumption that the initial source momentum is due solely to the previous beta emission, it is interesting to speculate about the energy available to the Xe^{131} atom following Coulomb fragmentation. Let us ignore the recoil from the preceding beta emission and assume the following sequence of events for the elemental iodine source:

$$I^{131} \rightarrow (IXe^{131})^+ \xrightarrow{\Delta t'} I^+ + Xe^{131+}. \qquad (2)$$

The ionization of the iodine partner could occur by means of a direct collision, by the sudden change in the electron-cloud configuration, or by some other as yet undetermined mechanism. A simple calculation based on the mutual repulsion of two singly charged ions indicates that in a time Δt ($\approx 4\times10^{-13}$ sec and corresponding to a separation distance of five times the initial molecular radius) a separation speed of 3.6×10^5 cm/sec will be reached. Translating these quantities into maximum effective Doppler shifts for the 637- and 723-keV transitions yields the results shown in Table II. Estimated maximum Doppler shifts for infinite separation are also given in Table II.

For the Coulomb fragmentation to be effective the requisite initial nuclear momentum must be attained in a time before the emission of the gamma ray. Referring to the postulated sequence of events shown in Eq. (2), one sees that this time is given by $\Delta t' + \Delta t$, where Δt is the ion separation time and $\Delta t'$ is the time interval between the beta decay and the rupturing of the molecular bond which initiates the Coulomb fragmentation. In this work the fraction of gamma rays emitted in a 1-eV interval having the correct energy to be resonantly scattered, $N(E_\gamma)/N$, for the 723-keV transition appeared to be at least three times greater than that for the 637-keV transition. If the $\Delta t'$ can be considered as the lifetime of a metastable or virtual state of the $(IXe)^+$ molecular ion, a value on the order of 10^{-12} sec would be consistent with our findings. The total time available would then allow a greater relative number of the 723-keV gamma rays to be resonantly scattered than would be the case

Table II. Comparison of the estimated Doppler shifts obtainable from Coulomb fragmentation with the recoil Doppler shifts for the three levels of interest in Xe^{131}. $r_0 \approx$ initial molecular radius and r is the separation distance of the ions.

E_γ (keV)	Coulomb fragmentation Doppler shift (eV) $r \rightarrow 5r_0$	Coulomb fragmentation Doppler shift (eV) $r \rightarrow \infty$	Recoil Doppler shift $2R_a$ (eV)
364	2.2	2.4	1.1
637	3.8	4.3	3.3
723	4.3	4.8	4.3

for gamma rays from the 637-keV level which, since it quite probably has a shorter lifetime,[7] would presumably suffer a reduction in the anticipated $N(E_\gamma)/N$ because of the number of photons emitted before the necessary initial nuclear momentum had been attained.

Although the actual molecular processes involved in providing the additional initial nuclear-recoil energy are undoubtedly much more complicated, the simple picture presented here is consistent with the gross features of the experimental results. At present, further experiments are under way in an attempt to learn more about the details of Coulomb fragmentation following beta decay and to apply this approach to other cases where otherwise one would not expect to be able to observe resonant scattering.

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ISOTOPE SHIFT IN THE 2^1S_0-2^1P_1 LINE OF HELIUM

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The isotope shift in the 2^1S_0-2^1P_1 (2.06-μ) line of helium has been measured for the first time and found to be 4200 ± 140 MHz. This measurement is a sensitive test of the mass-polarization contribution to the isotope shift and is found to be in excellent agreement with the calculations of Pekeris.

In recent years Pekeris[1] and his associates have calculated wave functions for the low-lying levels of the helium atom with great precision. A sensitive test of the accuracy of these results is the measurement of the isotope shift of the He^3-He^4 spectral lines. The isotope shift in the singlet spectra has two contributions, the reduced-mass effect and the mass-polarization term.[2] The reduced-mass effect gives rise to a shift in the energy level from E_∞, the Schrödinger solution for an infinitely heavy nucleus, to $E = [M/(M+m)]E_\infty$, where m and M are the electronic and nuclear masses, respectively. The mass-polarization term contributes a shift to the energy of $-(m/M)\int \psi_\infty \nabla_1 \cdot \nabla_2 \psi_\infty$ (atomic units) where ∇_1 and ∇_2 are the gradient operators for electrons one and two. This term is inversely proportional to the nuclear mass and dependent on the state of the atom, requiring accurate wave functions for its evaluation. Other contributions to E_∞ such as relativistic corrections are also calculated in Ref. 1, but these do not contribute to the isotope shift.

The isotope corrections calculated from Ref. 1 are shown in Fig. 1, where δE_{rm} and δE_{mp} denote, respectively, the reduced-mass and mass-polarization shifts in the energy. The reduced-mass terms were calculated directly from the He^4 data by use of the readily derived expressions

$$\delta E_{\mathrm{rm}}(\mathrm{He}^4) = \frac{m}{M_4}\nu,$$

$$\delta E_{\mathrm{rm}}(\mathrm{He}^3) = \frac{m}{M_3}\frac{M_3}{M_3+m}\left(\frac{M_4}{M_4+m}\right)^{-1}\nu,$$

where m is the mass of the electron, M_3 and M_4 the He^3 and He^4 nuclear masses, and ν the nonrelativistic ionization energy for the He^4 state, as given in Ref. 1 in wave numbers. The mass-polarization terms were obtained by

$$\delta E_{\mathrm{rm}}(\mathrm{He}^4) = \epsilon_M \text{ and } \delta E_{\mathrm{mp}}(\mathrm{He}^3) = (M_4/M_3)\epsilon_M,$$

where ϵ_M is the mass polarization correction as given in Ref. 1 in wave numbers. The masses expressed in unified nuclidic mass units were