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Observation of the uranium 235 nuclear magnetic resonance signal (*)

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Résumé. — La première observation du signal de résonance magnétique nucléaire de l'isotope 235 de l'uranium est présentée. Elle a été effectuée sur l'hexafluorure d'uranium pur, à l'état liquide à 380 K. Le rapport gyromagnétique mesuré est égal à $|\gamma(^{235}\text{U})| = 492,6 \pm 0,2 \text{ rad.s}^{-1} \text{ G}^{-1}$.

Abstract. — The first observation of the nuclear magnetic resonance of the uranium 235 is reported. It has been performed on pure liquid uranium hexafluoride at 380 K. The measured magnetogyric ratio is $|\gamma(^{235}\text{U})| = 492.6 \pm 0.2 \text{ rad.s}^{-1} \text{ G}^{-1}$.

The uranium 235 magnetic moment μ_N has previously been estimated only by indirect methods, the paramagnetic resonance of the U^{3+} ion inserted in a lanthanum chloride matrix [1, 2] and the optical absorption of the ^{235}U atom [2]. Both these methods indicate unambiguously that the nuclear spin is $I = 7/2$, but they only provide the magnetic moment value through delicate assumptions about quantities like $\langle r^{-3} \rangle$, where r is the electron spin to nuclear spin distance, the brackets corresponding to a quantum average of the relevant wave functions. The values of $|\mu_N|$ deduced thereof are found between 0.31 and 0.35 β_N , β_N being the nuclear magneton. These values, among the smallest of the periodic table, correspond to a magnetogyric ratio of 424 to 479 $\text{rad.s}^{-1} \text{ G}^{-1}$. The nuclear resonance frequency calculated from these values is between 1.58 % and 1.79 % that of the proton in the same magnetic field. On the other hand, the quadrupole moment of uranium 235 is very large, being estimated to 4.1 barns [1] or 6.4 barns [2]. Under these conditions, a very low sensitivity of detection is expected, explaining possibly why the NMR observation of this nucleus has not been reported before.

We report here the first NMR observation of ^{235}U . The uranium hexafluoride has been chosen for these experiments because the quadrupole effects are minimized by its cubic symmetry in the liquid state. Pure liquid UF_6 , 93.5 % enriched in ^{235}U , has been studied at 380 K, in a 10 mm o.d., 6 mm i.d. sealed silica tube. At this temperature, the UF_6 vapour pressure is ca. 4 bars.

The NMR spectra were recorded on a BRUKER WM 500 spectrometer with an external field of 11.747 T. The rf. power corresponds to a $\pi/2$ pulse of 90 μs . On the figure are compared the absorption spectra of UF_6 93.5 % enriched in ^{235}U (A), of UF_6 with a natural abundance of 0.72 % in this isotope (B) and an off resonance blank recording (C).

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Fig. 1. — (A) NMR spectrum of 93.5 % isotopically enriched $^{235}\text{UF}_6$, origin of the spectrum 9.126 666 MHz. (B) Spectrum of natural UF_6 , same origin as (A). (C) Off resonance baseline, origin : 9.336 666 MHz.

The measured resonance frequency of ^{235}U is $\nu = 9.209_8 \pm 0.004$ MHz, corresponding therefore to a magnetogyric ratio :

$$|\gamma(^{235}\text{U})| = (0.018\,41_4 \pm 0.000\,01) \gamma_{\text{H}} = 492.6 \pm 0.2 \text{ rad. s}^{-1} \text{ G}^{-1},$$

γ_{H} being the proton magnetogyric ratio in tetramethylsilane ($26\,751 \text{ rad. s}^{-1} \text{ G}^{-1}$). The nuclear moment given by the expression $|\mu_{\text{N}}| = |\gamma(^{235}\text{U})| \hbar I$ is thus equal to $0.359\,9_9 \pm 0.000\,15$ nuclear magnetons, with likely a negative sign according to reference 3. The μ_{N} value determined here is therefore very close to the largest one given in reference 2. Dorain *et al.* [2] indicate however that their estimate of $\langle r^{-3} \rangle$ is quite inaccurate being uncertain by a factor 2. Moreover, the electronic structures of the investigated species (U^{3+} , U and UF_6) are very different so that chemical effects of a few per cent may be expected for the corresponding effective nuclear magnetic moments.

The accuracy in our determination of μ_N is limited by two factors : a marked distortion of the base line by acoustic ringing [6] and a half height line width of 20 kHz of the ^{235}U signal.

In several articles, Ursu *et al.* [7-9] carried on an indirect study of the nuclear magnetism of ^{235}U through the relaxation of ^{19}F spins in UF_6 . These studies showed a dependence of the ^{19}F relaxation properties upon the isotopic composition of uranium, which was assigned to the relaxation of ^{235}U . Although the large value of the ^{235}U line width measured here is an evidence of an efficient quadrupolar relaxation mechanism induced by molecular vibrations, a quantitative comparison seems premature without complementary experiments. Among them an improvement of experimental conditions of observation of the ^{235}U signal (rf. power, pulse sequences) is needed. They might allow, in particular, a practical method for determining the uranium isotope composition, the most abundant one, ^{238}U , being devoided of nuclear spin.

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