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Newly identified energy levels and calculated transition rates in astatine atom, the rarest element on Earth

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ABSTRACT

A theoretical investigation of the atomic structure and radiative parameters involving the lowest states within the $6p^5$, $6p^47s$ and $6p^47p$ configurations of neutral astatine is reported for the first time in the present paper. Using different physical models based on the pseudo-relativistic Hartree–Fock approach, the influence of intravalence, core-valence and core-core electron correlation on the atomic parameters is discussed in detail. This work allowed us to establish the intermediate-coupling composition and to clearly fix the spectroscopic designation of six excited levels within the $6p^47s$ and $6p^47p$ configurations which had been located in previous experimental studies. In addition new tentative identifications of four levels belonging to the $6p^49p$ and $6p^410p$ configurations are reported in this work together with predicted level energies along the $6p^4np$ and $6p^4nd$ Rydberg series up to $n=50$.

1. Introduction

In view of the high radioactivity of all its isotopes, astatine ($Z=85$) is the rarest naturally occurring element on the Earth's crust with an estimated total abundance of less than one gram at any given time [1]. This element was discovered in 1940 by Corson et al. [2] by bombarding a bismuth target with α particles. As described by Fry and Thoennessen [3], thirty-nine astatine isotopes with atomic masses ranging from $A=191$ to $A=229$ have been discovered so far and, according to the Hartree–Fock–Bogoliubov (HFB-14) mass model [4], about 37 additional astatine isotopes could exist. All of these isotopes are characterized by very short half-lives, the most stable species being ^{210}At with a half-life time of only 8.1 h. This isotope's primary decay mode is positron emission to the relatively long-lived and very toxic ^{210}Po. In total, only five isotopes of astatine have half-lives exceeding one hour, namely those between $A=207$ and $A=211$. The least stable ground state isotope is ^{213}At, with a half-life of about 125 ns. It undergoes α decay to the extremely long-lived isotope ^{209}Bi. With a half-life of 7.2 h, the ^{211}At isotope is the subject of ongoing research in nuclear medicine since it decays either via emission of an α particle (to ^{207}Bi) or via electron capture (to an extremely short-lived nuclide, ^{211}Po, which undergoes further α decay). This makes ^{211}At an ideal short-range radiation source for targeted α therapy in cancer treatment (see e.g. [5–10]).

Because of its extreme rarity, the chemical and physical properties of astatine are almost totally unknown. Many of these have only been estimated based on its position in the periodic table, making of At the heaviest of halogens, i.e. the group of elements including fluorine, chlorine, bromine and iodine. As regards its electronic structure, the ground state of the astatine atom is $6p^5\ ^2P_{3/2}$ while the lowest excited configurations are of the type $6p^4nl$ (with $nl=7s$, 7p, 6d, 5f, ...). Nevertheless, only two excited levels ($6p^47s\ ^4P_{3/2,5/2}$) were clearly classified until now [11], although some additional tentative identifications of higher-lying states were suggested from recent experiments [12,13]. As support to further experimental investigations of astatine, it is however useful to know the atomic structure of this element in the most complete possible way. Heavy neutral atoms such as At I represent also an interesting challenge for the theoretical calculations in view of their very complex atomic structures containing a large number of electrons.

In the present work, different theoretical models based on the pseudo-relativistic Hartree–Fock (HFR) approach have been used to compute the atomic structure and radiative parameters involving the lowest states within the $6p^5$, $6p^47s$ and $6p^47p$ configurations with the aim of classifying the spectral lines and the energy levels observed in recent experiments. A tentative identification of higher states belonging to $6p^49p$ and $6p^410p$ configurations is also reported. This study can be considered as an extension of our previous theoretical investigations dedicated to the atomic properties of some other short-lived radioelements such as technecium ($Z=43$) [14], promethium ($Z=61$) [15], polonium ($Z=84$)

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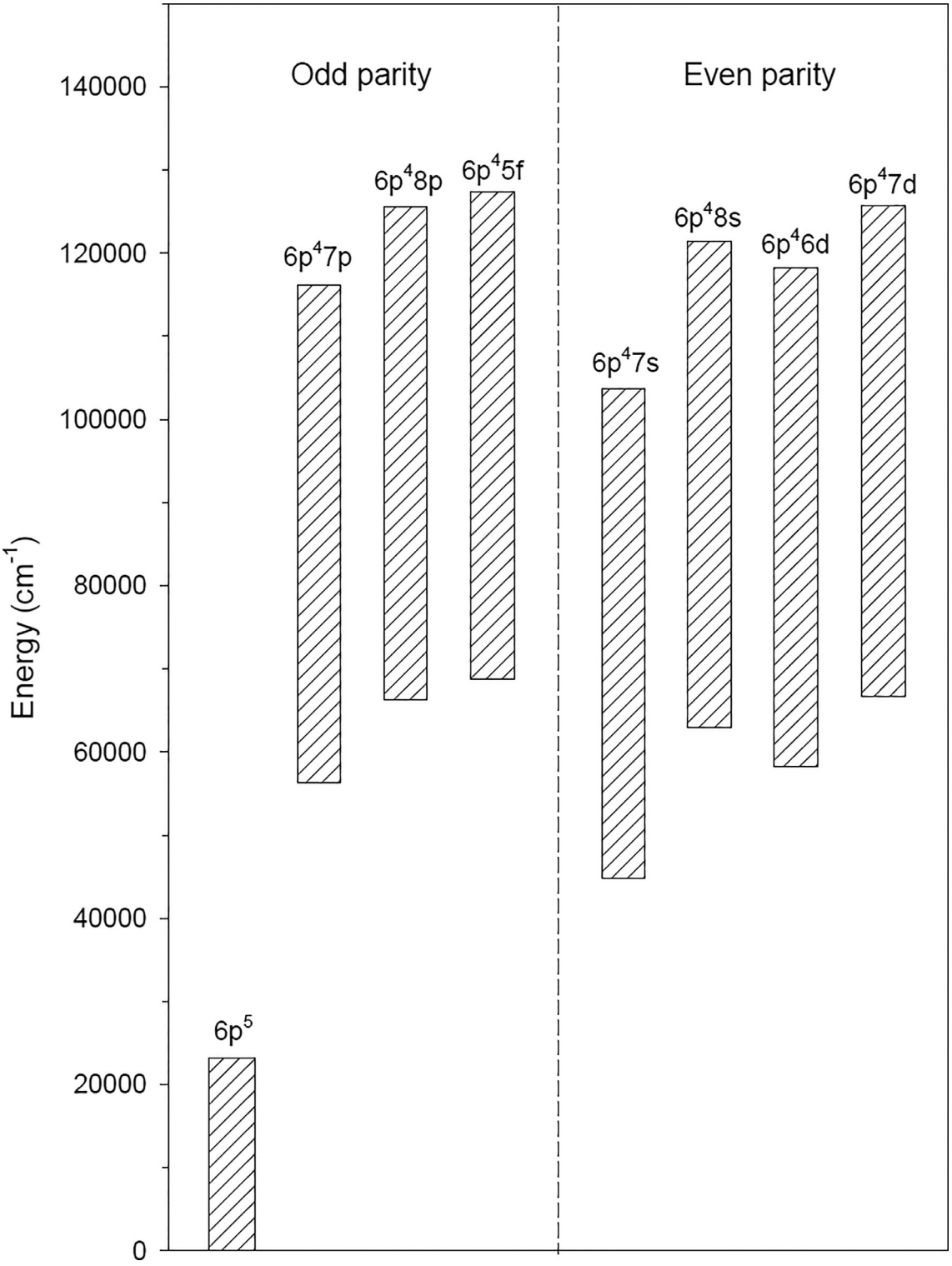

Fig. 1. Predicted locations of the lowest odd- and even-parity configurations in astatine atom.

[16], francium ($Z=87$) [17], radium ($Z=88$) [18], actinium ($Z=89$) [18] and americium ($Z=95$) [19].

2. Available experimental data

The first experimental study of the electronic structure of astatine atom was performed in 1964 by McLaughlin [11] who studied a 70 ng sample of artificially produced radioactive isotopes and used a highly sensitive method of spectrographic detection of gases to observe the At I absorption spectrum. Two lines were recorded whose wavelengths were 216.225 and 224.401 nm. These lines were tentatively assigned to the transitions from the ground state $6p^5\ ^2P_{3/2}$ to the even-parity energy levels $6p^4(^3P)7s\ ^4P_{3/2}$ and $6p^4(^3P)7s\ ^4P_{5/2}$ which were thus located at 46,233.6 and 44,549.3 cm^{-1}, respectively. Fifty years later, these tentative assignments were confirmed by in-source laser resonance ionization