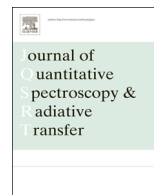




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## Investigation of intravalence, core–valence and core–core electron correlation effects in polonium atomic structure calculations

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### ABSTRACT

A detailed investigation of the atomic structure and radiative parameters involving the lowest states within the  $6p^4$ ,  $6p^36d$ ,  $6p^37s$ ,  $6p^37p$  and  $6p^37d$  configurations of neutral polonium is reported 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 fix the spectroscopic designation of some experimental level energy values and to provide for the first time a set of reliable oscillator strengths corresponding to 31 Po I spectral lines in the wavelength region from 175 to 987 nm.

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### 1. Introduction

Because of the high radioactivity of all its isotopes, polonium ( $Z=84$ ) is a very rare element in nature. According to Fry and Thoennessen [1], forty-two polonium isotopes with atomic masses ranging from  $A=186$  to  $A=227$  have been discovered so far. All of these isotopes are characterized by short half-lives, the most stable species being  $^{209}\text{Po}$  with a half-life of 102 years. Among naturally occurring polonium,  $^{210}\text{Po}$  has the longest half-life with 138.39 days. This isotope can be found in small concentrations in the earth's crust and in the atmosphere. Nevertheless, despite its rarity in our environment, this chemical element arouses a growing interest since a few years because of its potential applications in different scientific fields such as nuclear physics and astrophysics.

In nuclear physics, the spectroscopic properties of polonium has attracted much attention over the last years. In particular, laser-induced fluorescence spectroscopy in a collimated atomic beam was applied by Kowalewska and coworkers [2] to determine isotope shifts and the hyperfine structure of the isotopic chain  $^{200}\text{Po}$ ,  $^{202}\text{Po}$ ,  $^{204-210}\text{Po}$ . The measurements were based on the observation of resonance fluorescence after excitation of the atomic transition  $6p^4\ ^3P_2 \rightarrow 6p^37s\ ^5S_2$  of Po I. More recently, three new ionization schemes for polonium were tested with the resonant ionization laser ion source (RILIS) during the on-line production of  $^{196}\text{Po}$  in a  $\text{UC}_x$  target at CERN-ISOLDE. The saturation of the atomic transitions were observed and the yields of the isotope chain  $^{193-198,200,202,204}\text{Po}$  were measured [3]. With the same facility, in-source resonant ionization laser spectroscopy of the even- $A$  polonium isotopes  $^{192-210,216,218}\text{Po}$  was performed using the  $6p^37s\ ^5S_2 \rightarrow 6p^37p\ ^5P_2$  transition in Po I [4]. In this latter work, the comparison of the measured isotope shifts in  $^{200-210}\text{Po}$  with previous data set allowed the authors to test for the first time large-scale atomic calculations [5] used to extract the changes in the mean-square charge radius

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of the atomic nucleus. The same experiment was also used to extract new information on the  $\beta$  decay and the hyperfine structure of  $^{199}\text{Po}$  [6]. As support to further experimental or theoretical studies of nuclear properties in polonium, it is essential to know the atomic structure of this element with the highest possible accuracy.

In astrophysics, recent observations have suggested the presence of short-lived radioactive elements, such as Tc ( $Z=43$ ), Pm ( $Z=61$ ) and  $84 \leq Z \leq 99$  elements at the surface of the chemically peculiar roAp star HD 101065, also known as Przybylski's star [7–10]. In particular, Gopka et al. [8] presented the results on new identifications of the lines of all radioactive elements with atomic numbers from  $Z=84$  to  $Z=99$ , except for  $Z=85$  (At) and  $Z=87$  (Fr). The presence of these heavy short-lived radioactive elements in HD 101065 star is enigmatic (see discussion in [7–13]). Goriely [14] has stressed the importance of spallation nucleosynthesis compared to diffusion processes as a possible explanation of the chemical composition of the outer layers of some chemically peculiar stars. The available astrophysical spectroscopic observations are however affected by large uncertainties essentially due to the lack of atomic data for the elements of interest.

These heavy elements are 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, we concentrate on the particular case of neutral polonium (Po I) for which different pseudo-relativistic Hartree–Fock (HFR) models have been used to compute the atomic structure and radiative parameters involving the lowest states within the  $6p^4$ ,  $6p^36d$ ,  $6p^37s$ ,  $6p^37p$  and  $6p^37d$  configurations. This study is an extension of our previous theoretical works dedicated to the atomic properties of some other short-lived radioelements such as technetium ( $Z=43$ ) [15], promethium ( $Z=61$ ) [16], francium ( $Z=87$ ) [17], radium ( $Z=88$ ) [18], actinium ( $Z=89$ ) [18] and americium ( $Z=95$ ) [19].

## 2. The experimental spectrum and energy levels of polonium

In the current atomic spectra database available on the National Institute of Standards and Technology (NIST) website [20], 32 energy level values of Po I ranging from 0 to  $64451 \text{ cm}^{-1}$  are listed and classified as belonging to  $6p^4$ ,  $6p^37p$ ,  $6p^38p$ ,  $6p^39p$ ,  $6p^310p$  and  $6p^36d$ ,  $6p^37d$ ,  $6p^38d$ ,  $6p^37s$ ,  $6p^38s$  even-parity and odd-parity configurations, respectively. These values were essentially taken from the work of Charles [21] who revised and extended the previous experimental analyses of the polonium spectrum performed by Charles et al. [22] and Mrozowski [23]. More precisely, Charles [21] reported the wavelengths of 97 lines of  $^{208}\text{Po}$  and the hyperfine structure of 31 lines of  $^{209}\text{Po}$  covering the spectral range 210–900 nm obtained by measurement of Fabry–Perot interferograms using a radiation source containing a sample of polonium composed of 33 micrograms of  $^{208}\text{Po}$  and 11 micrograms of  $^{209}\text{Po}$ . From these data, relative values of 26 energy levels of  $^{208}\text{Po}$  I were deduced. However, for about half of those levels, the author was unable to give a spectroscopic designation or

even a total angular momentum  $J$ -quantum number with certainty. As an example, the three lowest energy levels of  $6p^37p$  configuration appearing at 50681.28, 50934.89 and  $51636.42 \text{ cm}^{-1}$  were reported by Charles [21] with uncertain  $J$ -values. However, more recently, three new ionization schemes for polonium were tested with the resonant ionization laser ion source (RILIS) during the on-line production of  $^{196}\text{Po}$  in a  $\text{UC}_x$  target at CERN-ISOLDE [3]. In the first ionization scheme, one valence electron was excited from the  $6p^4 \ ^3P_2$  ground state with a UV transition at 255.8 nm to the  $6p^37s \ ^5S_2$  excited state. A second excited state was reached with an infrared transition at 843.38 nm before the electron occupying this level was excited to the continuum by the non-resonant absorption of a 510.6 nm photon, provided by a Cu vapor laser. During this experiment, the resonance curve of the second step was observed confirming the existence of the excited state at  $50934.89 \text{ cm}^{-1}$  with  $6p^3(^4S)7p \ ^5P_2$  as spectroscopic designation.

## 3. Atomic structure calculations

The computational procedure that we have used for modeling the atomic structure and calculating radiative parameters in Po I is the pseudo-relativistic Hartree–Fock method described in detail by Cowan [24]. In order to investigate the effects of intravalence, core–valence and core–core electron correlations, six sets of calculations (A–F) were performed in the present work. These are summarized in Table 1. In view of the large number of uncertainties affecting the positions, the spectroscopic designations and/or the  $J$ -quantum numbers in the configurations with  $n \geq 7$ , we focused our study on the lowest states belonging to  $6p^4$ ,  $6p^36d$ ,  $6p^37s$ ,  $6p^37p$  and  $6p^37d$  configurations. As seen from Table 1, more and more electron correlation is progressively included when going from model A to model F. More precisely, in calculation A, only valence correlation outside  $6s^26p^3$  is considered while some single and double excitations from the  $6p$  subshell are included in calculations B and C, respectively. In calculation D, core–valence correlation effects are added by including single excitations from the  $6s$  subshell while, in calculation E, core–core electron correlation contributions are considered with double excitations from  $6s$ . Finally, in model F, the core-polarization (CPOL) effects from the 78 electrons occupying the  $1s^22s^22p^63s^23p^63d^{10}4s^24p^64d^{10}4f^{14}5s^25p^65d^{10}$  closed shells corresponding to the Po VII ionic core were estimated using a pseudo-potential and a correction to the dipole operator according to the HFR+CPOL procedure described in many of our previous papers (see e.g. [25–27]). These CPOL corrections depend on two parameters, i.e. the electric dipole polarizability of the ionic core,  $\alpha_d$ , and the cut-off radius,  $r_c$ , which can be seen as a measure of the size of the ionic core. For the first parameter, a value of  $\alpha_d = 2.00a_0^3$  was graphically extrapolated for Po VII from the theoretical values of Fraga et al. [28] along the platinum isoelectronic sequence (see Fig. 1). The cut-off radius,  $r_c$ , was chosen to be equal to  $1.17a_0$  which corresponds to the HFR average value  $\langle r \rangle$  for the outermost core orbital (5d).

**Table 1**  
Interacting configurations and core-polarization parameters retained in the HFR atomic structure calculations of neutral polonium.

Type of calculation	A	B	C	D	E	F	
Even configurations	6s <sup>2</sup> 6p <sup>4</sup> 6s <sup>2</sup> 6p <sup>3</sup> 7p 6s <sup>2</sup> 6p <sup>3</sup> 5f 6s <sup>2</sup> 6p <sup>3</sup> 6f	6s <sup>2</sup> 6p <sup>4</sup> 6s <sup>2</sup> 6p <sup>3</sup> 7p 6s <sup>2</sup> 6p <sup>3</sup> 5f 6s <sup>2</sup> 6p <sup>3</sup> 6f 6s <sup>2</sup> 6p <sup>2</sup> 6d <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 7s <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 6d7s 6s <sup>2</sup> 6p <sup>2</sup> 6d7d 6s <sup>2</sup> 6p <sup>2</sup> 7s7d 6s <sup>2</sup> 6p <sup>2</sup> 7p5f 6s <sup>2</sup> 6p <sup>2</sup> 7p6f	6s <sup>2</sup> 6p <sup>4</sup> 6s <sup>2</sup> 6p <sup>3</sup> 7p 6s <sup>2</sup> 6p <sup>3</sup> 5f 6s <sup>2</sup> 6p <sup>3</sup> 6f 6s <sup>2</sup> 6p <sup>2</sup> 6d <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 7s <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 6d7s 6s <sup>2</sup> 6p <sup>2</sup> 6d7d 6s <sup>2</sup> 6p <sup>2</sup> 7s7d 6s <sup>2</sup> 6p <sup>2</sup> 7p5f 6s <sup>2</sup> 6p <sup>2</sup> 7p6f 6s <sup>2</sup> 6p7p <sup>3</sup> 6s <sup>2</sup> 6p6d <sup>2</sup> 7p 6s <sup>2</sup> 6p7s <sup>2</sup> 7p 6s <sup>2</sup> 6p6d7s7p	6s <sup>2</sup> 6p <sup>4</sup> 6s <sup>2</sup> 6p <sup>3</sup> 7p 6s <sup>2</sup> 6p <sup>3</sup> 5f 6s <sup>2</sup> 6p <sup>3</sup> 6f 6s <sup>2</sup> 6p <sup>2</sup> 6d <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 7s <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 6d7s 6s <sup>2</sup> 6p <sup>2</sup> 6d7d 6s <sup>2</sup> 6p <sup>2</sup> 7s7d 6s <sup>2</sup> 6p <sup>2</sup> 7p5f 6s <sup>2</sup> 6p <sup>2</sup> 7p6f 6s <sup>2</sup> 6p7p <sup>3</sup> 6s <sup>2</sup> 6p6d <sup>2</sup> 7p 6s <sup>2</sup> 6p7s <sup>2</sup> 7p 6s <sup>2</sup> 6p6d7s7p 6s6p <sup>4</sup> 6d 6s6p <sup>4</sup> 7s 6s6p <sup>3</sup> 6d7p 6s6p <sup>3</sup> 7s7p	6s <sup>2</sup> 6p <sup>4</sup> 6s <sup>2</sup> 6p <sup>3</sup> 7p 6s <sup>2</sup> 6p <sup>3</sup> 5f 6s <sup>2</sup> 6p <sup>3</sup> 6f 6s <sup>2</sup> 6p <sup>2</sup> 6d <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 7s <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 6d7s 6s <sup>2</sup> 6p <sup>2</sup> 6d7d 6s <sup>2</sup> 6p <sup>2</sup> 7s7d 6s <sup>2</sup> 6p <sup>2</sup> 7p5f 6s <sup>2</sup> 6p <sup>2</sup> 7p6f 6s <sup>2</sup> 6p7p <sup>3</sup> 6s <sup>2</sup> 6p6d <sup>2</sup> 7p 6s <sup>2</sup> 6p7s <sup>2</sup> 7p 6s <sup>2</sup> 6p6d7s7p 6s6p <sup>4</sup> 6d 6s6p <sup>4</sup> 7s 6s6p <sup>3</sup> 6d7p 6s6p <sup>3</sup> 7s7p 6p <sup>6</sup> 6p <sup>5</sup> 7p 6p <sup>4</sup> 6d <sup>2</sup> 6p <sup>4</sup> 7s <sup>2</sup> 6p <sup>4</sup> 7p <sup>2</sup> 6p <sup>4</sup> 6d7s	6s <sup>2</sup> 6p <sup>4</sup> 6s <sup>2</sup> 6p <sup>3</sup> 7p 6s <sup>2</sup> 6p <sup>3</sup> 5f 6s <sup>2</sup> 6p <sup>3</sup> 6f 6s <sup>2</sup> 6p <sup>2</sup> 6d <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 7s <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 6d7s 6s <sup>2</sup> 6p <sup>2</sup> 6d7d 6s <sup>2</sup> 6p <sup>2</sup> 7s7d 6s <sup>2</sup> 6p <sup>2</sup> 7p5f 6s <sup>2</sup> 6p <sup>2</sup> 7p6f 6s <sup>2</sup> 6p7p <sup>3</sup> 6s <sup>2</sup> 6p6d <sup>2</sup> 7p 6s <sup>2</sup> 6p7s <sup>2</sup> 7p 6s <sup>2</sup> 6p6d7s7p 6s6p <sup>4</sup> 6d 6s6p <sup>4</sup> 7s 6s6p <sup>3</sup> 6d7p 6s6p <sup>3</sup> 7s7p 6p <sup>6</sup> 6p <sup>5</sup> 7p 6p <sup>4</sup> 6d <sup>2</sup> 6p <sup>4</sup> 7s <sup>2</sup> 6p <sup>4</sup> 7p <sup>2</sup> 6p <sup>4</sup> 6d7s	6s <sup>2</sup> 6p <sup>4</sup> 6s <sup>2</sup> 6p <sup>3</sup> 7p 6s <sup>2</sup> 6p <sup>3</sup> 5f 6s <sup>2</sup> 6p <sup>3</sup> 6f 6s <sup>2</sup> 6p <sup>2</sup> 6d <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 7s <sup>2</sup> 6s <sup>2</sup> 6p <sup>2</sup> 6d7s 6s <sup>2</sup> 6p <sup>2</sup> 6d7d 6s <sup>2</sup> 6p <sup>2</sup> 7s7d 6s <sup>2</sup> 6p <sup>2</sup> 7p5f 6s <sup>2</sup> 6p <sup>2</sup> 7p6f 6s <sup>2</sup> 6p7p <sup>3</sup> 6s <sup>2</sup> 6p6d <sup>2</sup> 7p 6s <sup>2</sup> 6p7s <sup>2</sup> 7p 6s <sup>2</sup> 6p6d7s7p 6s6p <sup>4</sup> 6d 6s6p <sup>4</sup> 7s 6s6p <sup>3</sup> 6d7p 6s6p <sup>3</sup> 7s7p 6p <sup>6</sup> 6p <sup>5</sup> 7p 6p <sup>4</sup> 6d <sup>2</sup> 6p <sup>4</sup> 7s <sup>2</sup> 6p <sup>4</sup> 7p <sup>2</sup> 6p <sup>4</sup> 6d7s
Odd configurations	6s <sup>2</sup> 6p <sup>3</sup> 7s 6s <sup>2</sup> 6p <sup>3</sup> 6d 6s <sup>2</sup> 6p <sup>3</sup> 7d	6s <sup>2</sup> 6p <sup>3</sup> 7s 6s <sup>2</sup> 6p <sup>3</sup> 6d 6s <sup>2</sup> 6p <sup>3</sup> 7d 6s <sup>2</sup> 6p <sup>2</sup> 6d7p 6s <sup>2</sup> 6p <sup>2</sup> 7s7p 6s <sup>2</sup> 6p <sup>2</sup> 6d5f 6s <sup>2</sup> 6p <sup>2</sup> 6d6f 6s <sup>2</sup> 6p <sup>2</sup> 7s5f 6s <sup>2</sup> 6p <sup>2</sup> 7s6f	6s <sup>2</sup> 6p <sup>3</sup> 7s 6s <sup>2</sup> 6p <sup>3</sup> 6d 6s <sup>2</sup> 6p <sup>3</sup> 7d 6s <sup>2</sup> 6p <sup>2</sup> 6d7p 6s <sup>2</sup> 6p <sup>2</sup> 7s7p 6s <sup>2</sup> 6p <sup>2</sup> 6d5f 6s <sup>2</sup> 6p <sup>2</sup> 6d6f 6s <sup>2</sup> 6p <sup>2</sup> 7s5f 6s <sup>2</sup> 6p <sup>2</sup> 7s6f 6s <sup>2</sup> 6p6d <sup>3</sup> 6s <sup>2</sup> 6p6d <sup>2</sup> 7s 6s <sup>2</sup> 6p6d7s <sup>2</sup> 6s <sup>2</sup> 6p6d7p <sup>2</sup> 6s <sup>2</sup> 6p7s7p <sup>2</sup>	6s <sup>2</sup> 6p <sup>3</sup> 7s 6s <sup>2</sup> 6p <sup>3</sup> 6d 6s <sup>2</sup> 6p <sup>3</sup> 7d 6s <sup>2</sup> 6p <sup>2</sup> 6d7p 6s <sup>2</sup> 6p <sup>2</sup> 7s7p 6s <sup>2</sup> 6p <sup>2</sup> 6d5f 6s <sup>2</sup> 6p <sup>2</sup> 6d6f 6s <sup>2</sup> 6p <sup>2</sup> 7s5f 6s <sup>2</sup> 6p <sup>2</sup> 7s6f 6s <sup>2</sup> 6p6d <sup>3</sup> 6s <sup>2</sup> 6p6d <sup>2</sup> 7s 6s <sup>2</sup> 6p6d7s <sup>2</sup> 6s <sup>2</sup> 6p6d7p <sup>2</sup> 6s <sup>2</sup> 6p7s7p <sup>2</sup> 6s6p <sup>5</sup> 6s6p <sup>4</sup> 7p 6s6p <sup>3</sup> 6d <sup>2</sup> 6s6p <sup>3</sup> 7s <sup>2</sup> 6s6p <sup>3</sup> 7p <sup>2</sup> 6s6p <sup>3</sup> 6d7s	6s <sup>2</sup> 6p <sup>3</sup> 7s 6s <sup>2</sup> 6p <sup>3</sup> 6d 6s <sup>2</sup> 6p <sup>3</sup> 7d 6s <sup>2</sup> 6p <sup>2</sup> 6d7p 6s <sup>2</sup> 6p <sup>2</sup> 7s7p 6s <sup>2</sup> 6p <sup>2</sup> 6d5f 6s <sup>2</sup> 6p <sup>2</sup> 6d6f 6s <sup>2</sup> 6p <sup>2</sup> 7s5f 6s <sup>2</sup> 6p <sup>2</sup> 7s6f 6s <sup>2</sup> 6p6d <sup>3</sup> 6s <sup>2</sup> 6p6d <sup>2</sup> 7s 6s <sup>2</sup> 6p6d7s <sup>2</sup> 6s <sup>2</sup> 6p6d7p <sup>2</sup> 6s <sup>2</sup> 6p7s7p <sup>2</sup> 6s6p <sup>5</sup> 6s6p <sup>4</sup> 7p 6s6p <sup>3</sup> 6d <sup>2</sup> 6s6p <sup>3</sup> 7s <sup>2</sup> 6s6p <sup>3</sup> 7p <sup>2</sup> 6s6p <sup>3</sup> 6d7s 6p <sup>3</sup> 6d 6p <sup>3</sup> 7s 6p <sup>4</sup> 6d7p 6p <sup>4</sup> 7s7p	6s <sup>2</sup> 6p <sup>3</sup> 7s 6s <sup>2</sup> 6p <sup>3</sup> 6d 6s <sup>2</sup> 6p <sup>3</sup> 7d 6s <sup>2</sup> 6p <sup>2</sup> 6d7p 6s <sup>2</sup> 6p <sup>2</sup> 7s7p 6s <sup>2</sup> 6p <sup>2</sup> 6d5f 6s <sup>2</sup> 6p <sup>2</sup> 6d6f 6s <sup>2</sup> 6p <sup>2</sup> 7s5f 6s <sup>2</sup> 6p <sup>2</sup> 7s6f 6s <sup>2</sup> 6p6d <sup>3</sup> 6s <sup>2</sup> 6p6d <sup>2</sup> 7s 6s <sup>2</sup> 6p6d7s <sup>2</sup> 6s <sup>2</sup> 6p6d7p <sup>2</sup> 6s <sup>2</sup> 6p7s7p <sup>2</sup> 6s6p <sup>5</sup> 6s6p <sup>4</sup> 7p 6s6p <sup>3</sup> 6d <sup>2</sup> 6s6p <sup>3</sup> 7s <sup>2</sup> 6s6p <sup>3</sup> 7p <sup>2</sup> 6s6p <sup>3</sup> 6d7s 6p <sup>3</sup> 6d 6p <sup>3</sup> 7s 6p <sup>4</sup> 6d7p 6p <sup>4</sup> 7s7p	6s <sup>2</sup> 6p <sup>3</sup> 7s 6s <sup>2</sup> 6p <sup>3</sup> 6d 6s <sup>2</sup> 6p <sup>3</sup> 7d 6s <sup>2</sup> 6p <sup>2</sup> 6d7p 6s <sup>2</sup> 6p <sup>2</sup> 7s7p 6s <sup>2</sup> 6p <sup>2</sup> 6d5f 6s <sup>2</sup> 6p <sup>2</sup> 6d6f 6s <sup>2</sup> 6p <sup>2</sup> 7s5f 6s <sup>2</sup> 6p <sup>2</sup> 7s6f 6s <sup>2</sup> 6p6d <sup>3</sup> 6s <sup>2</sup> 6p6d <sup>2</sup> 7s 6s <sup>2</sup> 6p6d7s <sup>2</sup> 6s <sup>2</sup> 6p6d7p <sup>2</sup> 6s <sup>2</sup> 6p7s7p <sup>2</sup> 6s6p <sup>5</sup> 6s6p <sup>4</sup> 7p 6s6p <sup>3</sup> 6d <sup>2</sup> 6s6p <sup>3</sup> 7s <sup>2</sup> 6s6p <sup>3</sup> 7p <sup>2</sup> 6s6p <sup>3</sup> 6d7s 6p <sup>3</sup> 6d 6p <sup>3</sup> 7s 6p <sup>4</sup> 6d7p 6p <sup>4</sup> 7s7p
Core-polarization parameters						$\alpha_d = 2.00a_0^3$ $r_c = 1.17a_0$ (Po VII core)	

In each of these approaches (from A to F), the final wavefunctions were obtained by a parametric fit of the calculated energy levels to the experimental values taken from [20,21]. More precisely, for the even parity, 8 level energies were used to optimize the average energy,  $E_{av}$ , the Slater electrostatic interaction integral,  $F^2(6p,6p)$ , the spin-orbit parameter,  $\zeta_{6p}$ , and the effective interaction parameter,  $\alpha$ , in the 6p<sup>4</sup> configuration as well as the average energy and the spin-orbit parameter,  $\zeta_{7p}$ , in the 6p<sup>3</sup>7p configuration. In the case of odd parity, 9 experimental energy levels were used to adjust the numerical values of

the average energy,  $E_{av}$ , the Slater direct,  $F^2(6p,6p)$ , and exchange,  $G^1(6p,7s)$ , integrals in 6p<sup>3</sup>7s configuration, together with the average energy,  $E_{av}$ , and the Slater exchange integrals,  $G^1(6p,6d)$  and  $G^3(6p,6d)$ , in 6p<sup>3</sup>6d configuration, with the constraint that the ratio between these two latter parameters is kept fixed during the fitting process. Note also that the 6p<sup>3</sup>7d average energy was adjusted using the experimental value of 6p<sup>3</sup>7d ( $J=2$ ) level at 59469.66 cm<sup>-1</sup> [21]. In addition, the ab initio HFR values for all the other Slater integrals,  $F^k$ ,  $G^k$  and  $R^k$ , not adjusted semi-empirically were scaled down by a

factor 0.80, as suggested by Cowan [24]. In Table 2, the lowest energy levels obtained below  $59,500\text{ cm}^{-1}$  using the most sophisticated model (F) are presented and compared with available experimental values. The main eigenvector components are also given for each level in the same table. In the work published by Charles [21], many levels were not identified by a full spectroscopic designation, some of them being even listed with an

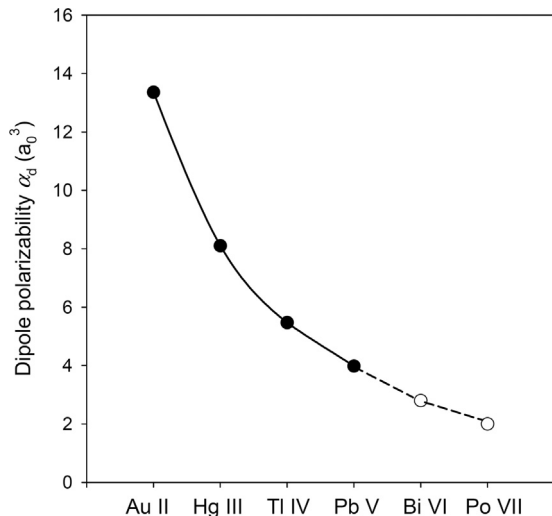


Fig. 1. Dipole polarizability along the platinum isoelectronic sequence. Full circles are theoretical values taken from [28]. Open circles connected with a dotted line represent extrapolated values.

Table 2

Lowly excited energy levels (below  $59,500\text{ cm}^{-1}$ ) in  $6p^4$ ,  $6p^36d$ ,  $6p^37s$ ,  $6p^37p$  and  $6p^37d$  configurations of Po I.

$E_{\text{exp}}$ ( $\text{cm}^{-1}$ ) <sup>a</sup>	$E_{\text{calc}}$ ( $\text{cm}^{-1}$ ) <sup>b</sup>	$\Delta E$ ( $\text{cm}^{-1}$ )	$J$	1st Component	2nd Component	3rd Component
<b>Even parity</b>						
0.00	0	0	2	78% $6p^4\ ^3P$	20% $6p^4\ ^1D$	
7514.69	7516	-1	0	55% $6p^4\ ^3P$	43% $6p^4\ ^1S$	
16831.61	16833	-1	1	98% $6p^4\ ^3P$		
21679.11	21680	-1	2	78% $6p^4\ ^1D$	20% $6p^4\ ^3P$	
42718.00	42717	1	0	54% $6p^4\ ^1S$	43% $6p^4\ ^3P$	
50681.28	50773	-92	1	34% $6p^3(^4S)7p\ ^5P$	16% $6p^3(^4S)7p\ ^3P$	15% $6p^3(^2P)7p\ ^3P$
50934.89	50838	97	2	40% $6p^3(^4S)7p\ ^5P$	15% $6p^3(^2P)7p\ ^3D$	13% $6p^3(^2P)7p\ ^3P$
51636.42	51641	-5	3	52% $6p^3(^4S)7p\ ^5P$	31% $6p^3(^2P)7p\ ^3D$	8% $6p^3(^2D)7p\ ^3F$
	51896		1	31% $6p^3(^4S)7p\ ^3P$	19% $6p^3(^4S)7p\ ^5P$	16% $6p^3(^2P)7p\ ^3S$
	52853		2	38% $6p^3(^4S)7p\ ^3P$	17% $6p^3(^2P)7p\ ^1D$	13% $6p^3(^2P)7p\ ^3P$
	53847		0	55% $6p^3(^4S)7p\ ^3P$	19% $6p^3(^2P)7p\ ^1S$	12% $6p^3(^2D)7p\ ^3P$
<b>Odd parity</b>						
39081.19	38837	244	2	52% $6p^3(^4S)7s\ ^5S$	32% $6p^3(^2P)7s\ ^3P$	9% $6p^3(^2D)7s\ ^3D$
40802.70	40551	252	1	43% $6p^3(^4S)7s\ ^3S$	22% $6p^3(^2P)7s\ ^1P$	20% $6p^3(^2D)7s\ ^3D$
51713.09	51846	-133	2	28% $6p^3(^4S)6d\ ^5D$	18% $6p^3(^4S)6d\ ^3D$	15% $6p^3(^2P)6d\ ^3D$
52098.93	52213	-114	3	47% $6p^3(^4S)6d\ ^5D$	17% $6p^3(^2P)6d\ ^3D$	14% $6p^3(^2P)6d\ ^3F$
52532.12	52375	157	1	44% $6p^3(^4S)6d\ ^5D$	17% $6p^3(^2P)6d\ ^3P$	12% $6p^3(^2P)6d\ ^3D$
	52835		4	53% $6p^3(^4S)6d\ ^5D$	31% $6p^3(^2P)6d\ ^3F$	8% $6p^3(^2D)6d\ ^3G$
53027.61	52863	165	2	26% $6p^3(^4S)6d\ ^3D$	21% $6p^3(^4S)6d\ ^5D$	6% $6p^3(^2D)6d\ ^3P$
	52904		0	54% $6p^3(^4S)6d\ ^5D$	30% $6p^3(^2P)6d\ ^3P$	7% $6p^3(^2D)6d\ ^3P$
	54000		3	46% $6p^3(^4S)6d\ ^3D$	20% $6p^3(^2P)6d\ ^1F$	11% $6p^3(^2D)6d\ ^3G$
54250.26	54316	-65	1	44% $6p^3(^4S)6d\ ^3D$	16% $6p^3(^2P)6d\ ^1P$	10% $6p^3(^2P)6d\ ^3P$
55923.80	56107	-183	2	37% $6p^3(^2D)7s\ ^3D$	36% $6p^3(^4S)7s\ ^5S$	16% $6p^3(^2D)7s\ ^1D$
57078.05	57321	-243	1	46% $6p^3(^2D)7s\ ^3D$	36% $6p^3(^4S)7s\ ^3S$	3% $6p^3(^4S)7d\ ^3D$
59469.66	59456	14	2	30% $6p^3(^4S)7d\ ^5D$	20% $6p^3(^4S)7d\ ^3D$	14% $6p^3(^2P)7d\ ^3D$

<sup>a</sup> Experimental energy levels from [21].

<sup>b</sup> Calculated energy levels as obtained in the present work using the HFR(F) model.

uncertain value of the  $J$ -quantum number. These gaps are now filled for the lowest excited levels of  $6p^4$ ,  $6p^37p$ ,  $6p^36d$ ,  $6p^37s$  and  $6p^37d$  configurations reported in Table 2. In particular, the three even-parity levels at 50,681.28, 50,934.89 and 51,636.42  $\text{cm}^{-1}$  were clearly identified in our calculations as belonging to the  $6p^3(^4S)7p\ ^5P$  multiplet with  $J=1, 2$  and 3, respectively, which seems to contradict the assumptions made by Charles [21] who suggested the values  $J=3?$ ,  $J=?$  and  $J=1$  or 2 for the same levels, respectively. However, it is worth noting that the spectroscopic designations obtained in the present work are confirmed by the same order of fine-structure levels observed within the  $5p^3(^4S)6p\ ^5P$  multiplet in the homolog Te I spectrum [20,29]. In the odd parity, the Po I levels at 54,250.26, 55,923.80 and 57,078.05  $\text{cm}^{-1}$ , not clearly identified by Charles [21] are now attributed to  $6p^3(^4S)6d\ ^3D_1$ ,  $6p^3(^2D)7s\ ^3D_2$  and  $6p^3(^2D)7s\ ^3D_1$ , respectively, the two latter states being however very strongly mixed with  $6p^3(^4S)7s\ ^5S_2$  and  $6p^3(^4S)7s\ ^3S_1$ .

#### 4. Radiative parameters

The theoretical oscillator strengths,  $\log gf$ , computed in the present work using the HFR models A–F described above are compared in Table 3 for 31 Po I lines involving all the experimentally known levels listed in Table 2. Note that transitions affected by severe cancellation effects in the calculation of the line strength, i.e. transitions for which the cancellation factor CF as defined by Cowan [24] is smaller than 0.01, were removed from the table.

**Table 3**Calculated oscillator strengths (log *gf*) for transitions in Po I using the different HFR models considered in the present work.

$\lambda$ (nm) <sup>a</sup>	Lower level (cm <sup>-1</sup> ) <sup>b</sup>		Upper level (cm <sup>-1</sup> ) <sup>b</sup>		A	B	C	D	E	F
175.199*	0.00	<i>J</i> =2 (even)	57078.05	<i>J</i> =1 (odd)	-1.02	-0.96	-1.02	-0.93	-0.87	-0.85
178.815*	0.00	<i>J</i> =2 (even)	55923.80	<i>J</i> =2 (odd)	-0.29	-0.30	-0.29	-0.38	-0.38	-0.36
184.331*	0.00	<i>J</i> =2 (even)	54250.26	<i>J</i> =1 (odd)	-0.78	-0.85	-0.84	-0.78	-0.79	-0.81
188.581*	0.00	<i>J</i> =2 (even)	53027.61	<i>J</i> =2 (odd)	-0.05	-0.11	-0.10	-0.15	-0.14	-0.15
190.360*	0.00	<i>J</i> =2 (even)	52532.12	<i>J</i> =1 (odd)	-1.61	-1.66	-1.61	-2.04	-1.96	-1.98
191.943*	0.00	<i>J</i> =2 (even)	52098.93	<i>J</i> =3 (odd)	-0.63	-0.76	-0.68	-0.63	-0.60	-0.61
201.697*	7514.69	<i>J</i> =0 (even)	57078.05	<i>J</i> =1 (odd)	-0.98	-1.07	-1.00	-1.08	-1.18	-1.19
213.9024	7514.69	<i>J</i> =0 (even)	54250.26	<i>J</i> =1 (odd)	-0.21	-0.25	-0.25	-0.25	-0.24	-0.26
222.0670	7514.69	<i>J</i> =0 (even)	52532.12	<i>J</i> =1 (odd)	-1.21	-1.27	-1.31	-1.27	-1.25	-1.26
234.4606	16831.61	<i>J</i> =1 (even)	59469.66	<i>J</i> =2 (odd)	-1.95	-1.84	-1.88	-1.78	-1.89	-1.87
245.0076	0.00	<i>J</i> =2 (even)	40802.70	<i>J</i> =1 (odd)	-0.06	-0.09	-0.08	-0.11	-0.11	-0.09
248.3942	16831.61	<i>J</i> =1 (even)	57078.05	<i>J</i> =1 (odd)	-0.23	-0.23	-0.24	-0.23	-0.23	-0.21
255.729*	16831.61	<i>J</i> =1 (even)	55923.80	<i>J</i> =2 (odd)	-1.16	-1.12	-1.15	-1.14	-1.16	-1.14
255.8008	0.00	<i>J</i> =2 (even)	39081.19	<i>J</i> =2 (odd)	-0.47	-0.46	-0.47	-0.46	-0.44	-0.42
264.5376	21679.11	<i>J</i> =2 (even)	59469.66	<i>J</i> =2 (odd)	-1.56	-1.51	-1.58	-1.46	-1.56	-1.54
276.1920	16831.61	<i>J</i> =1 (even)	53027.61	<i>J</i> =2 (odd)	-1.18	-1.23	-1.23	-1.13	-1.17	-1.21
280.0256	16831.61	<i>J</i> =1 (even)	52532.12	<i>J</i> =1 (odd)	-1.87	-1.96	-1.89	-2.05	-2.02	-1.96
282.4112	21679.11	<i>J</i> =2 (even)	57078.05	<i>J</i> =1 (odd)	-1.15	-1.17	-1.17	-1.15	-1.13	-1.11
286.6010	16831.61	<i>J</i> =1 (even)	51713.09	<i>J</i> =2 (odd)	-1.54	-1.56	-1.63	-1.72	-1.78	-1.80
291.9306	21679.11	<i>J</i> =2 (even)	55923.80	<i>J</i> =2 (odd)	-1.09	-1.13	-1.12	-1.13	-1.15	-1.14
300.3209	7514.69	<i>J</i> =0 (even)	40802.70	<i>J</i> =1 (odd)	-0.81	-0.82	-0.82	-0.81	-0.82	-0.80
306.9310	21679.11	<i>J</i> =2 (even)	54250.26	<i>J</i> =1 (odd)	-2.01	-2.04	-1.97	-2.34	-2.40	-2.36
324.0241	21679.11	<i>J</i> =2 (even)	52532.12	<i>J</i> =1 (odd)	-1.67	-1.72	-1.71	-1.88	-1.90	-1.85
328.6384	21679.11	<i>J</i> =2 (even)	52098.93	<i>J</i> =3 (odd)	-1.98	-1.96	-2.00	-2.00	-2.12	-2.14
417.0517	16831.61	<i>J</i> =1 (even)	40802.70	<i>J</i> =1 (odd)	-1.30	-1.40	-1.35	-1.42	-1.49	-1.49
574.4848	21679.11	<i>J</i> =2 (even)	39081.19	<i>J</i> =2 (odd)	-2.63	-2.63	-2.67	-2.52	-2.61	-2.62
696.184*	42718.00	<i>J</i> =0 (even)	57078.05	<i>J</i> =1 (odd)	-2.53	-2.57	-2.52	-2.60	-2.46	-2.46
796.2618	39081.19	<i>J</i> =2 (odd)	51636.42	<i>J</i> =3 (even)	0.57	0.54	0.54	0.53	0.53	0.52
843.3871	39081.19	<i>J</i> =2 (odd)	50934.89	<i>J</i> =2 (even)	0.32	0.27	0.28	0.27	0.28	0.27
861.8255	39081.19	<i>J</i> =2 (odd)	50681.28	<i>J</i> =1 (even)	0.02	-0.03	-0.03	-0.06	-0.06	-0.07
986.683*	40802.70	<i>J</i> =1 (odd)	50934.89	<i>J</i> =2 (even)	-0.40	-0.41	-0.41	-0.46	-0.43	-0.45

<sup>a</sup> Experimental wavelengths in vacuum (below 200 nm) and in air (above 200 nm) taken from [21]. Values with an asterisk \* were deduced from the experimental energy levels.

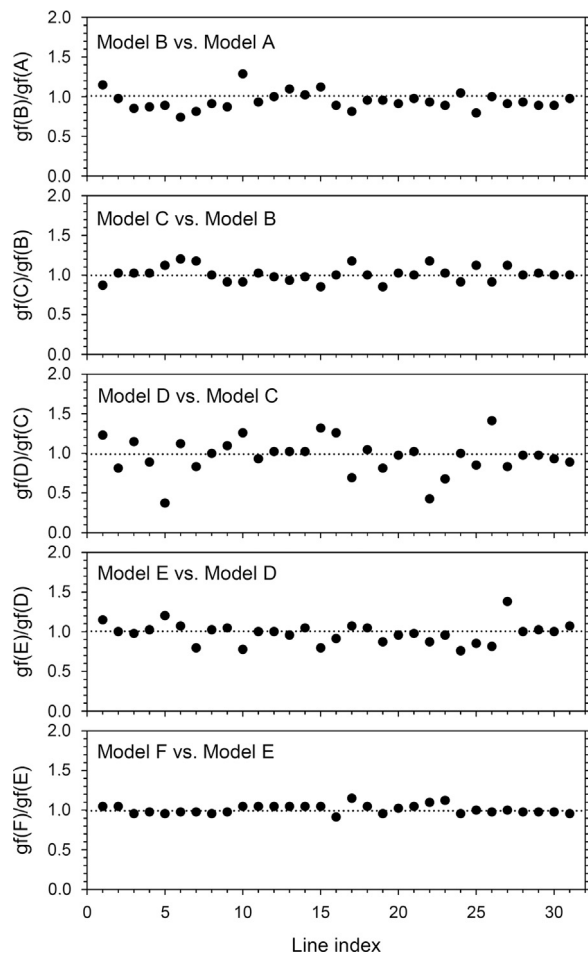
<sup>b</sup> Experimental energy level values taken from [21].

It is indeed well-known that the computed transition rates for such (usually weak) transitions may be expected to show very large errors. For information, these doubtful transition rates concern spectral lines situated at 168.153, 193.375, 267.167, 318.902, 332.860, 449.321, 522.769 and 866.895 nm.

The influence of the different types of electron correlation on oscillator strength calculations in polonium atom is illustrated in Fig. 2. In this figure, the ratios between *gf*-values computed using our different HFR models are compared for all the Po I lines listed in Table 3, numbered according to the order of appearance in that table. When looking at Fig. 2, one can observe that single and double excitations from both the 6s and 6p subshells (models B–E) play a non negligible role in the overall calculation of transition rates of interest while core-polarization effects from the Po VII [1s<sup>2</sup>...5d<sup>10</sup>] ionic core (model F) are rather small. This can be illustrated by the mean ratios of oscillator strengths obtained with two successive models for which the values were found to be equal to  $gf(B)/gf(A)=0.94 \pm 0.11$ ,  $gf(C)/gf(B)=1.01 \pm 0.09$ ,  $gf(D)/gf(C)=0.96 \pm 0.23$ ,  $gf(E)/gf(D)=0.98 \pm 0.13$  and  $gf(F)/gf(E)=1.01 \pm 0.05$  where the uncertainty represents the standard deviation.

However, when looking into more details, we note that the most important electron correlation effects on the

oscillator strengths depend on the type of transition considered. As an example, for the 6p<sup>4</sup>–6p<sup>3</sup>6d transitions, the most important changes in the *gf*-values are observed when considering models B and D. This is essentially because the 6s<sup>2</sup>6p<sup>2</sup>6d<sup>2</sup> and the 6s6p<sup>4</sup>6d configurations included in the 6s<sup>2</sup>6p<sup>4</sup> initial state in models B and D, respectively, have allowed dipole transitions to the 6s<sup>2</sup>6p<sup>3</sup>6d configuration. Furthermore, the radial dipole integral 6d→6p involved in the transition from the 6s<sup>2</sup>6p<sup>2</sup>6d<sup>2</sup> configuration has the same magnitude as the dipole matrix elements which connect the 6p<sup>4</sup> and 6p<sup>3</sup>6d main configurations. On the other hand, the equivalent configuration (6s<sup>2</sup>6p6d<sup>3</sup>) included in the 6s<sup>2</sup>6p<sup>3</sup>6d final state in model C does not have an allowed transition to the 6s<sup>2</sup>6p<sup>4</sup> initial state which explains why it is more important to include the 6p<sup>2</sup>→6d<sup>2</sup> double excitation in the initial than in the final state. Also the 6s→6d core excitation included in model D leads to a configuration (6s6p<sup>4</sup>6d) with an allowed dipole transition to the 6s<sup>2</sup>6p<sup>3</sup>6d final state which is not the case for the 6s<sup>2</sup>→6d<sup>2</sup> double excitation considered in model E. A similar discussion can be done concerning the influence of 6p<sup>2</sup>→7s<sup>2</sup> and 6s→7s core excitations on the oscillator strengths corresponding to the 6p<sup>4</sup>–6p<sup>3</sup>7s transitions. The situation is different for the 6p<sup>3</sup>7s–6p<sup>3</sup>7p transitions (i.e. the four last transitions in Table 3 and in Fig. 2). In this case, the computed oscillator strengths are only slightly sensitive



**Fig. 2.** Comparison between oscillator strengths obtained in the present work using different HFR models. In each panel, the y-axis gives the ratio of  $gf$ -values computed with two successive models, i.e. including an increasing number of interacting configurations (see text). The x-axis of the plots corresponds to the Po I spectral line indexes, which are assigned according to the order of appearance in Table 3 (i.e. by increasing wavelengths).

(within 10%) to single excitations from the  $6p$  (model B) and from the  $6s$  (model D) core subshells while double excitations from the same subshells have a negligible effect.

## 5. Conclusion

Different physical models based on the pseudo-relativistic Hartree-Fock method have been used for modeling the atomic structure and for computing radiative parameters in neutral polonium. They have emphasized the dominating influence of core-valence and core-core correlation for the  $6p^4-6p^36d$  and  $6p^4-6p^37s$  transitions. They also allowed us to provide a new set of decay rates for lines involving the lowest states in the  $6p^4$ ,  $6p^36d$ ,  $6p^37s$ ,  $6p^37p$  and  $6p^37d$  configurations and to give a reliable spectroscopic designation to some energy levels not clearly classified in previous works. This investigation is expected to provide a theoretical support to laser-spectroscopy experiments at ISOLDE and to check other

theoretical methods that will be used to compute hyperfine structure and isotope shift electronic parameters in the future. In addition, the new set of oscillator strengths obtained in the present work provides the basic information for a quantitative analysis of the polonium content in some chemically peculiar stars.

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