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Confinement, correlation and relativistic effects on the photoionization of 6s subshell of Hg

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Abstract. A theoretical study of photoionization parameters of Hg atom confined in a spherical shell potential has been carried out using the relativistic random phase approximation. It is found that confinement, correlation and relativistic effects all bring significant and separate changes in the 6s photoionization cross-section, and also in the photoelectron angular distribution.

1. Introduction

Confined quantum systems such as an atom trapped in fullerene cage have stimulated a number of studies in physics, chemistry and in interdisciplinary areas like material science and nanoscience. The presence of an external potential has significant effects on the physical and chemical properties of the atom trapped inside [1, 2, 3]. Such studies lead to important applications in the areas of nano-structures, quantum dots etc.

In the case of Ca@C₆₀, the endohedral environment has been reported to bring out a significant redistribution of oscillator strength and also to make ‘electron-correlations’ act in a manner opposite to that in free atoms [4]. Though theoretical investigations on confined atoms are progressing very rapidly [5, 6, 7, 8], experimental studies are scanty because of the difficulties in producing such systems in large concentrations. However, this scenario is now changing; investigations on A@C₆₀ using ultraviolet, laser and/or synchrotron radiation are already under way [9, 10].

Recently it has been shown that fullerene cage can be effectively used to isolate an atom from its environment and can be used as a building block for q-bits in quantum computer [11]. Photoionization studies of endohedral atoms A@C₆₀, where A is an atom trapped inside the fullerene cage, have aroused considerable attention in the past few years [12, 13, 14, 15]. We report in the present paper that encapsulation of an atom inside a fullerene cage leads to oscillations in the photoionization cross-section, and also in the angular distribution parameters. These additional oscillations in the photoionization parameters are called ‘confinement resonances’ [12, 16].

In the case of free atoms, when relativistic effects are taken into account, the ‘ns’ angular distribution asymmetry parameter deviates [17, 18, 19] from ‘ $\beta = 2$ ’ near a ‘Cooper minimum’ [20]

because of the dynamical difference between the $s \rightarrow \epsilon p_{1/2}$ and $s \rightarrow \epsilon p_{3/2}$ transition channels. To the best of our knowledge, no study of combined effects of relativistic interactions and confinement on cross-section and angular distribution has been reported earlier.

2. Methodology

We have modeled endohedral environment by a spherical attractive annular potential $V(r)$ given by,

$$V(r) = \begin{cases} U_0 < 0, & \text{if } r_c \leq r \leq r_c + \Delta, \\ 0, & \text{otherwise,} \end{cases}$$

where $r_c = 5.8$ a.u. is the inner radius of the shell having a thickness $\Delta = 1.9$ a.u. [21]. The potential depth $U_0 = 8.22$ eV is obtained from the experimental data on C_{60} determined from the electron affinity parameter [22, 23].

We investigate the effect of confinement and relativistic interactions on the photoionization of 6s subshell of Hg using Relativistic Random Phase Approximation (RRPA) [24]. RRPA is based on Dirac equation and hence relativistic interactions are included *a-priori* explicitly. It includes certain types of correlations in the initial and final states to all orders. Using RRPA, photoionization process can be described by a set of coupled integro-differential equations similar in structure to that of Hartree-Fock equations. In the RRPA, length and velocity forms of the transition matrix elements are equal. For computational economy we have used a truncated RRPA by coupling dipole channels from 6s, 5d, 5p, 5s, 4f, 4d, 4p, 4s subshells only. The loss of gauge invariance resulting from truncation is nevertheless rather minor.

In order to understand the effect of relativistic interactions we have compared our RRPA results with those from Random Phase Approximation with Exchange (RPAE) method which does not include relativistic effects.

3. Results and Discussion

All figures in this manuscript are plotted against the photoelectron energy since the ionization thresholds for free and confined mercury are different. Figure 1 shows the 6s photoionization cross-section of free and confined Hg. In free Hg, the cross-section undergoes a Cooper minimum below 5 eV. As in the case of free Hg, the 6s cross-section in @Hg also goes through a Cooper minimum [17], although at a higher energy around 11eV.

For @Hg, the cross-section shows oscillations in the region above the Cooper minimum in both RRPA and RPAE. They are completely missing in free Hg. These confinement resonances are the result of interference between the outgoing photoelectron's wavefunction and that part of the wavefunction which is reflected by the annular potential well, U_0 . This is similar to the fine structures seen in X-ray absorption spectrum where the escaping electron gets scattered by the surrounding charge cloud [26-28].

The structure seen below 12 eV in RPAE curve is caused by the autoionization resonances arising from the interference of photoionization (bound \rightarrow continuum) channels from 6s and excitation (bound \rightarrow bound) channels from 5d subshells. The present RRPA algorithm is not designed to obtain detailed information of the resonance region. A detailed analysis of the resonance region can be done using a combination of RRPA in conjunction with Relativistic Multichannel Quantum Defect Theory (RMQDT) [25].

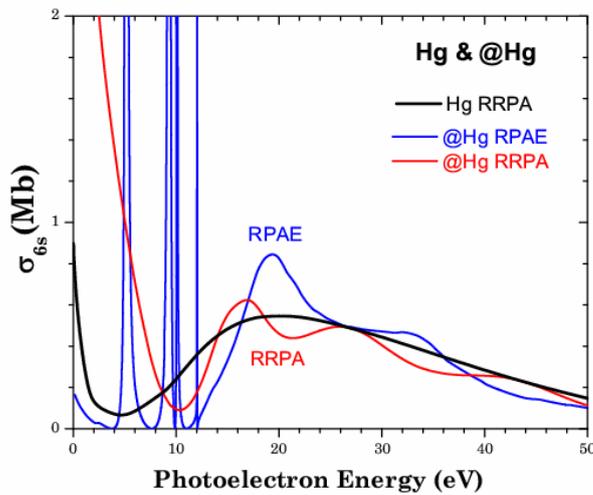


Figure 1 Photoionization cross-section of free Hg and @Hg

In the present work, focus is on the background cross-section for which we have used interpolation in the autoionization region. The photoionization cross-section maxima of the confinement resonances in RRPA occur at a lower energy than the position of the *corresponding maxima* in RPAE curve. This is obviously attributed to relativistic effects.

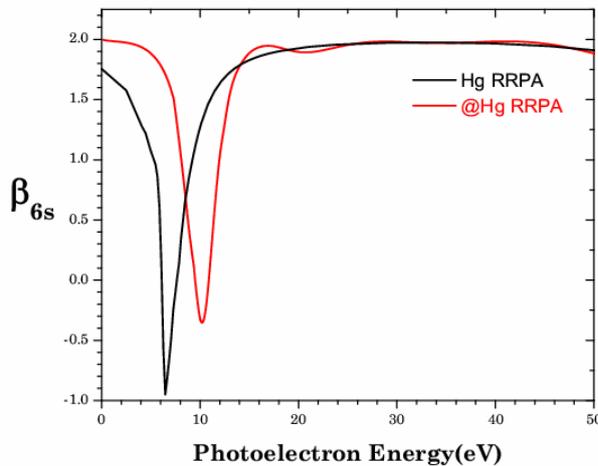


Figure 2 Angular distribution asymmetry parameter of free and confined Hg

In Figure 2 is shown the angular distribution asymmetry parameter β for Hg and for @Hg. In each case β deviates from 2.0 near the Cooper minimum. This phenomenon arises purely due to the effect of relativistic interactions since the two relativistic amplitudes $ns \rightarrow \epsilon p_{1/2}$ and $ns \rightarrow \epsilon p_{3/2}$ go through zero at different energies [29]. A detailed study of Hg 6s photoionization cross-section and angular distribution parameter is reported elsewhere [19]. *Combined* effects of relativistic interactions and confinement manifest as oscillations in the angular distribution asymmetry parameter and cause β to deviate from 2; departure from $\beta = 2$ is due to exchange of oscillator strengths in the two relativistic channels $ns \rightarrow \epsilon p_{1/2}$ and $ns \rightarrow \epsilon p_{3/2}$ whereas the oscillations are due to 'back-scattering' of the escaping photoelectron by the fullerene cage.

In order to understand the effect of interchannel coupling in @Hg, we have carried out a pilot study to include minimal interchannel coupling correlations nevertheless at two levels of truncation of the RRPA:

- (i) two dipole channels from 6s alone
- (ii) two dipole channels from 6s, three dipole channels from $5d_{5/2}$ and three E1 channels from $5d_{3/2}$.

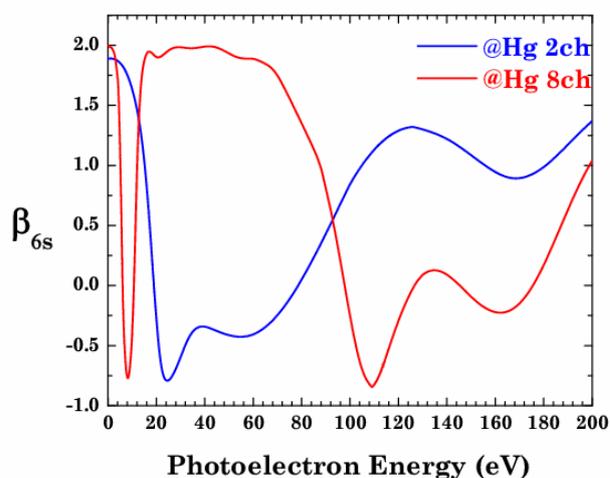


Figure 3 Angular distribution asymmetry parameter of @Hg at two different levels of truncation including channels from (i) 6s alone (ii) 6s and 5d.

In the case of free atomic Hg, inclusion of additional correlations induce multiple Cooper minima in the 6s photoionization [19]. In figure 3 is shown the angular distribution asymmetry parameter of @Hg for the pilot calculations mentioned above. In the photon energy region below the 5d thresholds we have in figure 3 presented background values of β_{6s} since this energy region contains sharp oscillations due to coupling between $6s \rightarrow \epsilon p$ ionization channels and $5d \rightarrow np, n'f$ excitation channels. In the case of @Hg interchannel coupling correlation effects are seen as in the case of free Hg. There is only a single Cooper minimum in the 2 channel calculation (dotted curve) seen ~ 24 eV. In 8ch calculation (solid curve) there are two Cooper minima, one which appears ~ 8 eV and another which appears ~ 109 eV. Additional oscillations seen above the Cooper minima are the confinement resonances due to the back-scattering of the escaping photoelectrons.

4. Conclusions

Relativistic interactions cause the value of 6s angular distribution asymmetry parameter β in @Hg to deviate from '2' near the vicinity of Cooper minima. The deviation of β at other energy points is attributed to the combined effects of relativistic interactions, confinement and interchannel coupling.

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