Application of coupled equation method on resonance processes of atomic lithium

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The coupled equation method (CEM) has been applied to investigating the resonance structures for the ground state $1s^2 2s\ ^2S$ of the neutral lithium from the first threshold up to 64.5 eV. Resonance structures of atomic lithium due to single excitations of the $1s$ and $2s$ electrons are studied by infinite-order calculations in detail. The effect of spin-orbit splitting is also included for some of the low-lying $1s2snp(\uparrow\downarrow)$ resonance, and the influence of the interference between $1s2s\ ^3Snp\downarrow$ and $1s2s\ ^1Snp\uparrow$ states on the resonance structure has been confirmed theoretically. The results show that the presented technique can give the reasonable resonance structures very well in photoionization processes.

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The study of photoionization processes of small atoms has been carried out recently by using accurate theoretical treatment. Therefore, a better understanding of both the electronic structure of target states and the dynamics of the resonance processes can be obtained. Lithium is one of a few metal elements for which there exists absolute absorption cross section data in a wide photon energy region[1−6]. This enables the theoretical models to be tested in a detailed manner[7−12]. Ederer *et al.*[5] first measured the K-shell photoabsorption spectrum of atomic lithium, using a synchrotron background source and a vapor furnace. The $1s^2 2s(^2S) \rightarrow 1s2s(^{3,1}S)np(^2P)$ resonance structures were also observed experimentally for incident photons in the energy range of $50-70$ eV. Ferrett *et al.*[2] performed measurements on $1s$-photoionization of lithium corresponding to the $1s2s^1S$ and ^3S ionic states. Langer *et al.*[3] also measured the partial cross sections and angular distribution asymmetry parameters of $1s$-photoionization using synchrotron radiation. However, there is almost no theoretical result of the photoionization of atomic lithium by using many-body perturbation theory (MBPT), especially in the $1s2s(^{3,1}S)np$ resonance region. Lithium photoionization has also been studied with a MBPT, but only at the edge of the non-resonant region between 5- and 10-eV photon energy[10]. MBPT has been proven to be very useful in the study of many-particle systems[3−15]. In the present work, the standard MBPT formulated by Brueckner[13] and Goldstone[14] and the extensions developed by Kelly[15] for application to atomic problems have been used to investigate the photoionization cross section of the ground state ^2S of the neutral lithium atom. In addition, we applied coupled equation method (CEM, essentially a close-coupling technique) to take account of the inter-channel interactions in the final state in order to obtain the accurate resonance structures.

This paper describes an investigation of the following processes related to $1s$ and $2s$ electrons ionization and excitation, in which the ionization system is left in the ^2P state:

$$h\nu + \text{Li}(1s^2 2s\ ^2S) \rightarrow \text{Li}^+(1s2s\ ^1S) + e^-(\epsilon p, np), \quad (1)$$

$$\rightarrow \text{Li}^+(1s2s\ ^3S) + e^-(\epsilon p, np), \quad (2)$$

$$\rightarrow \text{Li}^+(1s^2\ ^1S) + e^-(\epsilon p, np). \quad (3)$$

The calculations have demonstrated that the continuum configuration interaction has played an important role in the photon energy region of resonance. This inter-channel interaction was shown to transfer significant intensity from the direct channels into the conjugate shake-up channels, particularly for the final ionic states of the same multiplicity.

When one considers the interactions of the electrons in an atom with an electromagnetic radiation field polarized in the z direction, in the dipole approximation, the lowest-order contribution to photoionization cross section $\sigma(\omega)$ from the single-particle state Φ_a (denoted by $|a\rangle$) occupied in Φ_0 is given by[16]

$$\sigma(\omega) = (8\pi\omega/ck)|\langle r|z|a\rangle|^2, \quad (4)$$

where

$$k = (2\epsilon_a + 2\omega)^{1/2}, \quad (5)$$

and $\langle r|$ denotes the excited state, which does not occupy in Φ_0. In this paper, we use a, b, c, and d to be core orbitals and r, s, t and v to run over all the bound or continuum excited orbitals.

Therefore, one can calculate the cross section $\sigma(\omega)$ by evaluating the perturbation expansions of dipole transition matrix elements $\langle z \rangle_{ar} = \langle r|z|a\rangle$. These matrix may also be expressed by diagrams. The lowest-order contribution to $\sigma(\omega)$ or $\langle z \rangle$ of Eq. (4) is shown in Fig. 1(a). This diagram only represents the dipole matrix element $\langle r|z|a\rangle$. This is the ideal case. Actually, the electrons in an atom may interact with each other, and these interactions denoted by $\langle v \rangle_{rsab} = \langle rs|r^{-1}|ab\rangle$ can influence the dipole interaction matrix. The dashed lines with a dot at the end represent the dipole interaction and the solid lines without any ends represent the interaction correlation in Fig. 1. Each line with an arrow drawn upward denotes an occupied excited single particle state (a particle), and each line with an arrow drawn downward

denotes a vacancy in the initial state (a hole). In an actual many-electron atom as one know, many interactions between different excitation channels must be coupled with dipole matrix to infinite order, as shown in Fig. 2.
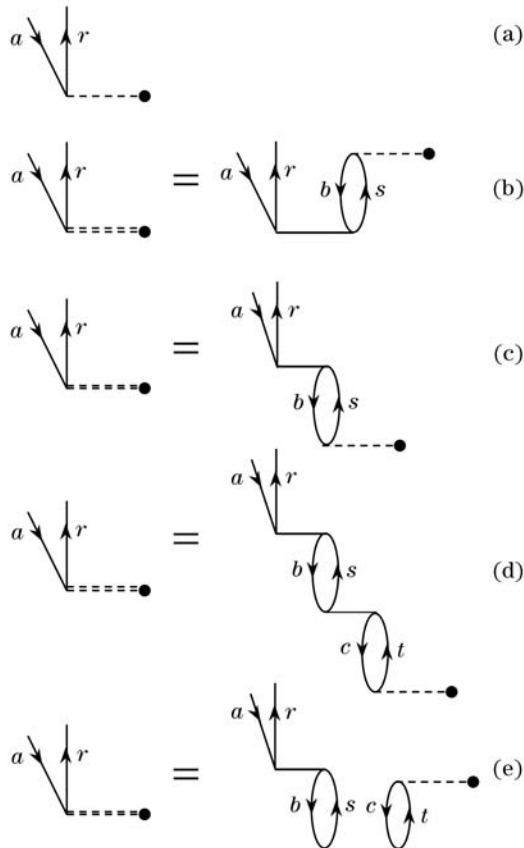

Fig. 1. The lowest- and first-order diagrams for single photoionization processes. Some second-order random phase approximation diagrams are also included.
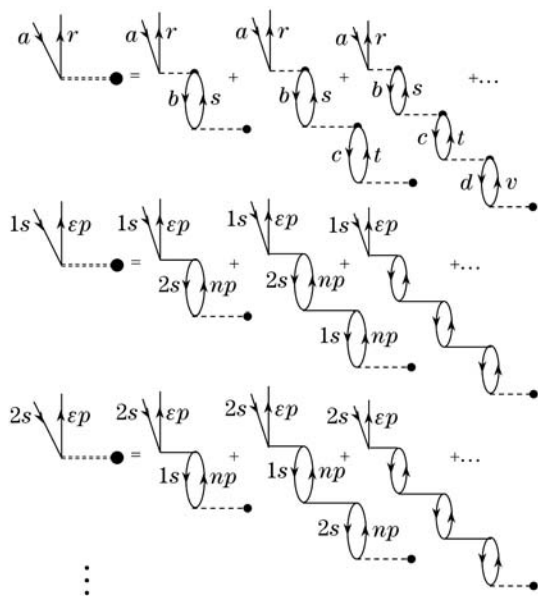

Fig. 2. Symbolic representation of the coupled equation for single photoionization process. The dashed line terminated with a dot represents the lowest-order dipole matrix element, the double dashed line terminated with a dot represents the correlated dipole matrix element.

The lowest-order dipole matrix element $\langle z \rangle$ in Eq. (4) should only be replaced by a correlated dipole matrix element $\langle \overline{z} \rangle$ in this case. The equations for these correlated dipole matrix elements then are coupled together.

The ground state orbitals in our basis were calculated by using Froese-Fischer's computer program MCHF[17]. We adopted the codes developed by Kelly[16] to calculate the continuum states and bound states respectively. Excitations of $1s$ and $2s$ electrons from the ground state $1s^2 2s(^2S)$ to the final states which have total orbital angular momentum $L = 1$ and total spin $S = 1/2$ were considered in calculations. Fourteen bound excited states and 56 continuum orbitals ranging from 0.05 to 5.80 a.u. were explicitly calculated for each l value. We report results of photoionization cross sections $\sigma(\omega)$ for the ground state ^2S of lithium with a configuration $1s^2 2s$. To gain a better understanding, we split the entire spectrum into two energy regions and made a detailed comparison with experiment in each region.

The resonance structures of photoionization cross sections are shown in Figs. 3 and 4, where the resonance positions are marked. The resonance energy is the position relative to the energy of the exact ground state. The threshold and resonance energy positions have been taken from the high-resolution photoelectron data of Langer et al.[3]. All of the resonances in the absorption cross section are consistent with that in Ref. [5]. A quantum-defect analysis[5] suggested that the resonance at 64.05 eV lying between the $[1s2s\ ^3S]6p$ and $[1s2s\ ^3S]7p$ resonances was
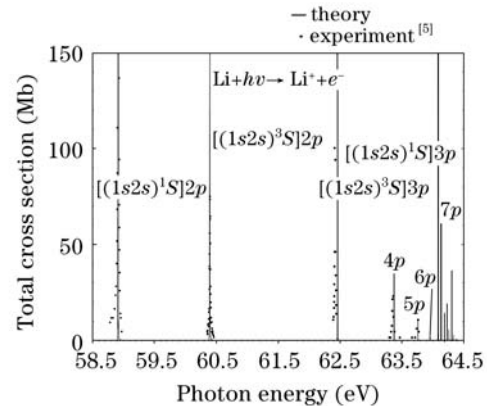

Fig. 3. Photoionization cross sections of atomic lithium below $1s2s\ ^3S$ threshold, $58.5 - 64.5$ eV region.
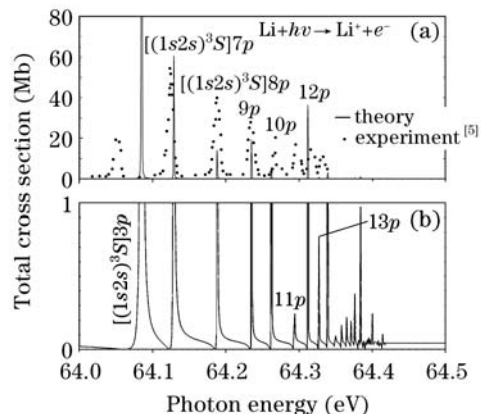

Fig. 4. Photoionization cross sections of atomic lithium below $1s2s\ ^3S$ threshold, $64 - 64.5$ eV region.

a perturber $[1s2s\ ^1S]3p$. It is well known that the Auger width of a Rydberg series, such as $[1s2s\ ^3S]np$, should decrease monotonically as n increases[10]. Our calculations show that there is also a strong intensity variation in the neighboring resonances of $[1s2s\ ^1S]3p$ at 64.09 eV as well as a quantum-defect shift confirming this suggestion.

In our data, the Rydberg series associated with the $[1s2s\ ^3S]np\ ^2P$ configuration are much more prominent, blend smoothly into the $1s2s\ ^3S\epsilon p$ continuum at 64.41 eV and are observable up to $n = 14$. Between the $[1s2s\ ^3S]6p$ and $[1s2s\ ^3S]7p$ members of this series there is a known perturber which has been studied by Chung[10], using the saddle-point technique, and is classified as the $[1s2s\ ^1S]3p\ ^2P$ state. The influence of the perturber on the intensity and energy position of both neighboring members of the $[1s2s\ ^3S]np\ ^2P$ series ($n = 6$ and 7) is clearly evident.

Immediately below the $1s2s\ ^3S$ threshold one can observe the higher members of $[1s2s\ ^3S]np\ ^2P$ Rydberg series in Fig. 4. The feature in the neighborhood of $1s2s\ ^3S$ threshold probably is not a resonance at all but part of the broad resonant structure above this limit. In the neighborhood of the $1s2s\ ^3S$ threshold the observed structure is due in part to the resonant structure above this threshold and in part to the higher member of the $[1s2s\ ^3S]np$ series which are not resolved[6].

Our calculations give excellent agreement with experiments and the previous theoretical calculations. The effectiveness of the present approach has been demonstrated by exploring the resonance structure in the photoionization cross sections of lithium. The availability of accurate experimental results for lithium enabled the theoretical models to be tested in a detailed way. The close agreement with the high-resolution measurements indicates the high accuracy of the present approach. The agreement appears to be excellent throughout the energy region from $1s^2\ ^1S$ threshold up to 64.5 eV.

In summary, one hand, this paper demonstrates that CEM can give reasonable resonance structure for atomic lithium by taking the infinite-order inter-channel interactions into account. On the other hand, the proposed approach and the related computational technique can be applied directly to more complex many-electron systems with a similar level of computational effect.

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