

## Radiation damping in the photoionization of Fe<sup>14+</sup>

M. F. Hasoglu,<sup>1,2</sup> T. W. Gorczyca,<sup>3</sup> M. A. Bautista,<sup>3</sup> Z. Felfli,<sup>4</sup> and S. T. Manson<sup>1</sup>

<sup>1</sup>Georgia State University, Atlanta, Georgia 30303, USA

<sup>2</sup>Gazikent University, 27100 Sahinbey, Gaziantep, Turkey

<sup>3</sup>Western Michigan University, Kalamazoo, Michigan 49008, USA

<sup>4</sup>Center for Theoretical Studies of Physical Systems, Clark Atlanta University, Atlanta, Georgia 30314, USA

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A theoretical investigation of photoabsorption and photoionization of Fe<sup>14+</sup> extending beyond an earlier frame transformation *R*-matrix implementation is performed using a fully correlated, Breit-Pauli *R*-matrix formulation including both fine-structure splitting of strongly bound resonances and radiation damping. The radiation damping of  $2p \rightarrow nd$  resonances gives rise to a resonant photoionization cross section that is significantly lower than the total photoabsorption cross section. Furthermore, the radiation-damped photoionization cross section is found to be in good agreement with recent experimental results once a global shift in energy of  $\approx -3.5$  eV is applied. These findings have important implications. First, the presently available synchrotron experimental data are applicable only to photoionization processes and not to photoabsorption; the latter is required in opacity calculations. Second, our computed cross section, for which the *L*-shell ionization threshold is aligned with the NIST value, shows a series of  $2p \rightarrow nd$  Rydberg resonances that are uniformly 3–4 eV higher in energy than the corresponding experimental profiles, suggesting that the actual *L*-shell threshold energy is lower than the value obtained using the current NIST data.

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Most of what we know about our universe is obtained through spectroscopy. We study either the emission of hot plasma sources or the absorption of intervening gas between us and a bright object. For example, it is possible to probe the multiphase interstellar medium (ISM) through the observation of atomic absorption lines and edges in the spectra of background sources, and the absorption properties of the plasma depend on the *photoabsorption* cross sections of the species present [1]. To interpret emission spectra, on the other hand, one first needs to determine the ionization structure of the gas, which, for photoionized plasmas, depends on the *photoionization* cross sections of the chemical elements present. Unlike spectra in the visual region dominated by valence electron processes in low-ionization species, extreme uv and x-ray spectra show inner *L*- and *K*-shell processes for which *photoionization* and *photoabsorption* are significantly different. The difference between these two processes arises from the fact that absorption of a photon by the atom to an autoionizing state (above the threshold) does not necessarily lead to electron emission.

For the present case of photons incident on Fe<sup>14+</sup> in the vicinity of the *L* edge, the difference between photoabsorption and photoionization can be understood by considering the prominent  $2p \rightarrow nd$  absorption resonances. Following absorption of a photon,

$$h\nu + 2p^63s^2 \longrightarrow 2p^53s^2nd, \quad (1)$$

an intermediate resonance state can either decay via autoionization, or Auger decay, leading to ionization of Fe<sup>14+</sup> into the Fe<sup>15+</sup> ion plus a free electron, or it can radiatively stabilize, leading to no charge change of the Fe<sup>14+</sup> ion:

$$2p^53s^2nd \longrightarrow 2p^63s + e^- \left( A_a^{\text{part}} \sim n^{-3} \right) \quad (2)$$

$$\longrightarrow 2p^6nd + e^- \left( A_a^{\text{spect}} \text{ ind. of } n \right) \quad (3)$$

$$\longrightarrow 2p^63s^2 + h\nu' \left( A_r^{\text{part}} \sim n^{-3} \right) \quad (4)$$

$$\longrightarrow 2p^63snd + h\nu' \left( A_r^{\text{spect}} \text{ ind. of } n \right). \quad (5)$$

Thus, the alternative decay pathways of radiative stabilization redirect some of the initial photoabsorption amplitude, giving a reduced, or *damped* [2], photoionization cross section relative to the photoabsorption cross section.

The extent of this so-called radiation damping [2] effect, for an isolated resonance, is essentially given by the autoionization branching ratio

$$\frac{\sigma_{PI}}{\sigma_{PA}} \approx \frac{A_a^{\text{tot}}}{A_a^{\text{tot}} + A_r^{\text{tot}}}, \quad (6)$$

where  $\sigma_{PI}$  and  $\sigma_{PA}$  are the photoionization and photoabsorption cross sections, respectively. The total autoionization and radiative rates,  $A_a^{\text{tot}}$  and  $A_r^{\text{tot}}$ , are each the sum of contributions for which the *nd* Rydberg electron *participates*, with partial rates scaling as  $n^{-3}$ , and for which the Rydberg electron is a *spectator*, with partial rates that are independent of *n*. Thus, the spectator Auger and radiative decay branches [Eqs. (3) and (5)] dominate as  $n \rightarrow \infty$ . On the other hand, due to angular momentum, or geometric, and radial, or dynamic, considerations of the *3d* orbital compared to the *3s* orbital, the  $2p^53s^23d$  resonance decays predominantly via participator pathways, as given in Eqs. (2) and (4).

To date, essentially all of the calculated photon-atom data for multiply charged ions has been produced by theoretical photoionization calculations without considering the alternate radiative decay pathway in Eqs. (4) and (5) (see, for example, Refs. [3,4]). These undamped cross sections are equivalent to the photoabsorption cross sections regarding absorption strength (although the resonance width is underestimated), and thus it had always been assumed that  $\sigma_{PI} = \sigma_{PA}$ . For Fe<sup>14+</sup>, perhaps the most definitive calculations to date are the *R*-matrix photoionization calculations in both *LS* coupling [5]

and using an  $LS$ -coupled to a  $JK$ -coupled frame transformation ( $LSJKFT$ ) method, along with resonance broadening in order to correctly characterize the resonances [6]. In that latter study (hereafter referred to as paper I), it was demonstrated that the  $LSJKFT$  method was capable of reproducing all fine-structure splitting effects of a full Breit-Pauli  $R$ -matrix (BPRM) calculation, except for the lowest  $2s^22p^53s^23d$  and  $2s2p^63s^23p$  resonances. This is because in the  $LSJKFT$  method, an extremely efficient computational approach compared to the BPRM method, the multichannel quantum defect (MQDT) equations for the outer-region solutions are modified, thereby incorporating fine-structure splitting only for those resonances that reside outside the  $R$ -matrix “box;” those that reside within the  $R$ -matrix region (i.e., all states described by orbitals only up to  $n = 3$ , such as the  $2s^22p^53s^23d$  resonances) did not include fine-structure splitting effects.

The agreement between  $LSJKFT$  and BPRM methods (except for the strongly bound resonances) was first demonstrated only for a minimal configuration-interaction (CI) case, since the large-scale CI calculation which would have been required for a more converged calculation and which was performed only within an  $LSJKFT$  approach, would have been prohibitively large to undertake within a full BPRM approach. Interestingly, it was found that in terms of general qualitative features, the large-CI  $LSJKFT$  cross section, convoluted with a sufficiently broad energy distribution, was essentially the same as that from a minimal-CI  $LS$  calculation. Thus, it is necessary to include higher-order CI and fine-structure effects only if a detailed resonance description is desired. It should be noted that all of those earlier calculations were for the total photoabsorption cross section and did not consider radiation damping effects. Furthermore, those earlier cross sections were all preconvoluted [7] with a constant Lorentzian width of 0.1 Ry [8], so a detailed investigation of the resonance profiles was not performed.

More recently, an electron-beam ion trap (EBIT) was used at a synchrotron facility to detect the final production of  $Fe^{15+}$  ions [9], thereby measuring the *photoionization* cross section at an energy resolution as low as 150 meV (see also Ref. [10]). Thus, it is now desirable to compute *radiation-damped* photoionization cross sections, since only total photoabsorption cross sections were reported in paper I. Furthermore, to compare directly with the finer resolution of the experimental data, it is necessary to consider higher-order CI effects and fine-structure splitting for all resonances, including the lowest  $2s^22p^53s^23d$  resonances for which a frame transformation approach is inapplicable. The present study improves upon the theoretical work of paper I and other earlier calculations [9,11] by using the same large-scale CI description of paper I within a full BPRM calculation, which treats fine-structure splitting correctly for even the lowest resonances, and by including radiation damping effects [12]. The BPRM calculations are performed using the standard  $R$ -matrix codes [2] with additional modifications for incorporating radiation damping [12], as described in earlier electron-impact excitation [13] and dielectronic recombination [14] studies. A brief description of the present methodology is given below, followed by a detailed comparison between these results and the recent experimental results.

The atomic structure used is essentially the same as that of paper I. The  $Fe^{15+}$  target states are described by an orbital basis set that is generated from a Hartree-Fock [15] calculation on the  $1s^22s^22p^63s$  ground-state configuration, with additional  $3p$  and  $3d$  orbitals generated from frozen-core Hartree-Fock calculations on the  $1s^22s^22p^63p$  and  $1s^22s^22p^63d$  excited configurations, respectively. All target states of the form  $2s^22p^63l$ ,  $2s^22p^53s3l$ , and  $2s2p^63s3l$  are used in the close-coupling expansion and are described by a larger CI basis consisting of all configurations consistent with single and double promotions out of those three base configurations.

The corresponding  $(N + 1)$ -electron bound, continuum, and resonance states of  $Fe^{14+}$  are described by a basis consisting of single, double, and triple promotions out of the  $2s^22p^63s^2$  base configuration. Lastly, unlike in paper I, corrections to the  $N$ -electron Hamiltonian are applied in order to realign the target energies with the recommended NIST [16] values, and the binding energy of the  $Fe^{14+}$  ground state, relative to the  $Fe^{15+}$  state, is also aligned with the NIST value. Thus, of particular importance, the present theoretical photon energy thresholds which we report are aligned with the corresponding NIST values.

The important details regarding radiation damping are that all  $2s^22p^63s^2$  and  $2s^22p^63s3d$  ( $J = 0, 1, 2$ ) bound states are included as final type II (inner-region) radiative decay states, resulting in a complex inner-region  $R$  matrix [12–14]. This accounts for all the participator radiative decay pathways in Eq. (4) and spectator radiative decay of the  $2p^53s^23d$  resonance. The spectator radiative decay of all higher  $nd$  resonances in Eq. (5), on the other hand, is accounted for by a modification to the MQDT equations in the outer region [12–14], where the type I ( $3s \rightarrow 2p$  core) decay width used in the  $E \rightarrow E - i\Gamma_r/2$  modification is  $\Gamma_r = 0.45$  meV.

The target states included explicitly in our  $R$ -matrix calculation only account for the participator Auger decay of Eq. (2) and the  $n = 3$  spectator Auger decay of Eq. (3) to the so-called *main-line* photoionization continua. The *satellite* photoionization continua of Eq. (3) for  $n > 3$  are instead included in our formulation via an optical potential MQDT modification procedure [17], similar to that done for the spectator radiative decay. This presents a difficulty in extracting a total theoretical photoionization cross section, since the above-described methodology for including radiation damping and  $n > 3$  spectator Auger decay yields only total photoabsorption and main-line photoionization cross sections [17]. The difference between the two includes both satellite photoionization and radiative damping amplitudes. As  $n \rightarrow \infty$ , the branching of the two can be extracted by using the spectator rates in Eq. (6), whereas for low  $n$ , the difference is due purely to radiation damping. For intermediate  $n$ , however, the branching requires a deeper investigation beyond the scope of the present work.

In Fig. 1, the present BPRM results are compared to the earlier results of paper I and to recent experiment [9]. In Fig. 1(a), our BPRM photoabsorption results, convoluted with a Lorentzian width of 0.1 Ry, are found to be qualitatively similar to the earlier  $LSJKFT$  results but differ quantitatively in three minor respects. First, there is a uniform energy difference between the two since the earlier results were computed using theoretical thresholds (NIST values were not available

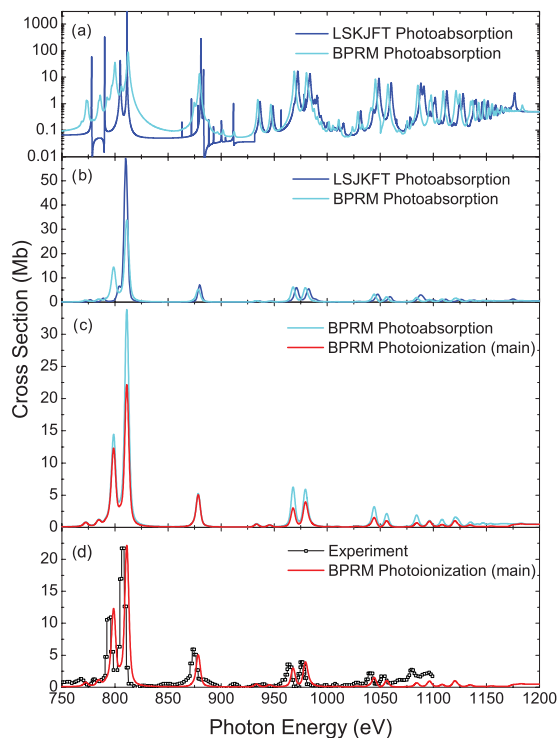


FIG. 1. (Color online)  $\text{Fe}^{14+}$  photoabsorption and main-line photoionization cross sections in the vicinity of the  $L$  edge. (a) Earlier  $LSJKFT$  photoabsorption results [6] (blue line) and present BPRM photoabsorption results (cyan line) using a Lorentzian width of 0.1 Ry. (b) Same as (a) using a Voigt profile ( $\Gamma_L = 2.11$  eV,  $\Gamma_G = 4.0$  eV). (c) Present BPRM photoabsorption results (cyan line) and main-line photoionization results (red line) using the same Voigt profile. (d) Present main-line photoionization results (red line) and experimental results [9] (black data points).

for the  $2p^53s^2$  autoionizing states at that time), whereas our present results have been aligned to the NIST thresholds. Second, the  $LSJKFT$  results were preconvoluted [7] with an 0.1 Ry Lorentzian [8] only above 931.55 eV, whereas the present BPRM results have preconvoluted even the lowest resonances. Third, and most importantly, the  $LSJKFT$  results do not account for fine-structure splitting of the lowest  $3d$  resonances, as noted earlier. The BPRM and  $LSJKFT$  cross sections are also compared to each other in Fig. 1(b) on a linear scale, convoluted with a Voigt profile (Lorentzian and Gaussian widths  $\Gamma_L = 2.11$  eV and  $\Gamma_G = 4.0$  eV, respectively). It is seen that, except for the slight global energy shift and the BPRM fine-structure splitting of the  $2s^22p^53s^23d$  resonances, the two cross sections are nearly identical. Thus, the only improvement over the  $LSJKFT$  method by using instead a BPRM method is the inclusion of fine-structure splitting of the lowest resonances, as had been noted in paper I.

Using the same resolution, we compare our total photoabsorption cross section to the computed main-line, damped photoionization cross section in Fig. 1(c). As noted above, the main-line photoionization cross section for the  $n = 3$  resonances below 950 eV photon energies, and, for the most part, the  $n = 4$  resonances, accounts for the total photoionization cross section, and thus there is appreciable  $3d \rightarrow 2p$  radiation

TABLE I.  $2p^53s^2(^2P_{1/2})3d$  resonance strengths (Mb eV).

Experiment [9]	$110 \pm 60$
$R$ -matrix photoionization	130
$R$ -matrix photoabsorption	200

damping of the lowest  $2p^53s^2nd$  resonances; the  $2s2p^63s^23p$  resonance at 880 eV, on the other hand, is not damped significantly due to the much smaller  $3p \rightarrow 2s$  radiative rate.

We compare the main-line photoionization cross sections to the experimental results (arbitrarily scaled) in Fig. 1(d). Except for an approximately constant energy difference, good agreement between the two results is found in resonance profiles up to about 1050 eV. It is interesting to note that the relative heights of the  $2p^53s^2(^2P_{3/2})3d$  and  $2p^53s^2(^2P_{1/2})3d$  resonances are the same in both the BPRM photoionization and experimental cross sections, whereas the BPRM photoabsorption cross section indicates a much different height ratio.

The comparison of relative  $2p^53s^23d$  resonance heights would seem to validate experimentally the extent of radiation damping. Nevertheless, we can get a more quantitative assessment by examining *absolute* resonance strengths. By fitting our photoionization and photoabsorption cross sections, in the vicinity of the  $2p^53s^2(^2P_{1/2})3d$  resonance, to an energy-normalized Lorentzian multiplied by a strength factor, we obtained integrated resonance strengths of 130 Mb eV and 200 Mb eV, respectively. Table I indicates that our photoionization resonance strength is indeed in agreement with the experimental value of  $110 \pm 60$  Mb eV, whereas the photoabsorption resonance strength is outside of the experimental uncertainty, providing further validation of the extent of radiation damping.

The comparison in Fig. 1(d) also indicates that the BPRM resonance energies are uniformly higher than the corresponding experimental values. This difference is quantified in Table II, which indicates that the resonance energy difference does not approach zero with the expected  $1/n^3$  behavior. The photon energy threshold values to which our calculations have been aligned—1172.8 eV for the  $2p^53s^2(^2P_{3/2})$  level and 1185.2 eV for the  $2p^53s^2(^2P_{1/2})$  level—are determined from the following NIST recommendations. First, the  $2p^63s^2 \rightarrow 2p^63s$  ionization energy is given by  $457.0 \pm 2.5$  eV [18], which is consistent with the value of  $456.2 \pm 0.5$  eV from the same EBIT experiment [9] and with an independent estimate of  $456.3 \pm 0.5$  eV [19]. Second, the  $2p^63s \rightarrow 2p^53s^2(^2P_{3/2,1/2})$  excitation energies are given by  $715.8 \pm 0.4$  eV and  $728.2 \pm 0.4$  eV, respectively. However, a more recent experimental measurement finds a value of  $713.8 \pm 0.4$  eV for the lower excitation [20]. Thus, while the overall experimental threshold uncertainties allow for the energy differences seen between theory and experiment in Table II, the fact that the differences do not approach zero as  $n$  increases, and the more recent excitation energy determination [20,21], both suggest that the true threshold energy is significantly lower than that used in the present calculations.

In conclusion, we have reported  $R$ -matrix calculations that improved on our earlier work of paper I by including fine-structure effects for the lowest resonance and radiation

TABLE II.  $\text{Fe}^{14+}$  resonance energies (eV). The theoretical resonance series are aligned to the  $2p^5 3s^2 ({}^2P_{3/2,1/2})$  thresholds given by NIST [16] (see text).

Level	Experimental [9]	Present	Difference
$\text{Fe}^{14+}(2p^5 3s^2 ({}^2P_{3/2})3d)$	$794.7 \pm 0.2$	798.8	4.1
$\text{Fe}^{14+}(2p^5 3s^2 ({}^2P_{3/2})4d)$	$964.3 \pm 0.2$	967.8	3.5
$\text{Fe}^{14+}(2p^5 3s^2 ({}^2P_{3/2})5d)$	$1040.9 \pm 0.5$	1044.2	3.3
$\text{Fe}^{14+}(2p^5 3s^2 ({}^2P_{1/2})3d)$	$807.1 \pm 0.2$	811.1	4.0
$\text{Fe}^{14+}(2p^5 3s^2 ({}^2P_{1/2})4d)$	$976.2 \pm 0.4$	979.3	3.1
$\text{Fe}^{14+}(2p^5 3s^2 ({}^2P_{1/2})5d)$	$1053.5 \pm 0.4$	1055.9	2.4

damping effects for all resonances. We find that radiation damping is significant for the  $2p^5 3s^2 ({}^2P_{1/2})3d$  resonance, in particular. Our radiation-damped photoionization resonance strengths show good agreement with the experimental results. However, there is a nearly uniform difference in energy positions between the present theoretical and the experimental [9] resonance positions. This discrepancy suggests that the correct threshold energies are lower than

those used in our calculations, which were obtained from the recommended NIST ionization and excitation energy values.

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