X-ray-absorption near-edge structure of laser-dressed neon

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Using an improved value for the energy splitting between the $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ states, electromagnetically induced transparency for x rays at the $1s \rightarrow 3p$ resonance in Ne is reassessed. The energy splitting between the $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ states is calculated with two independent approaches. In the first approach, first-order nonrelativistic many-body perturbation theory is employed. In the second approach, the Dirac-Fock method is combined with the relativistic configuration interaction method. Both approaches give an energy splitting of 1.88 eV. The calculated x-ray absorption near-edge structure of laser-dressed Ne agrees closely with an earlier prediction. It may therefore be expected that at a laser intensity of 10^{13} W/cm^2, the $1s \rightarrow 3p$ x-ray absorption cross section of Ne is suppressed by more than an order of magnitude.

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I. INTRODUCTION

The most prominent feature in the x-ray absorption cross section in the pre-edge region near the K edge of atomic neon is the resonance associated with the transition from the atomic ground state ($^1S_0$) to the $1s^{-1}3p\ ^1P_1$ state at an x-ray photon energy of 867.1 eV [1–6]. In Ref. [7], it was predicted that by exposing Ne atoms to a laser intensity of 10^{13} W/cm^2 at a wavelength of 800 nm, the x-ray absorption cross section at the $1s \rightarrow 3p$ resonance of Ne may be suppressed by a factor of 13. This phenomenon was termed "electromagnetically induced transparency for x rays" [7].

The physical basis for this effect is easy to understand. According to the Hartree-Slater-based atomic-structure model [8,9] employed in Ref. [7], the laser at 800 nm is in approximate resonance with the transition between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$. The $1s^{-1}3p\ ^1P_1$ state in the presence of N_L laser photons is thus degenerate with the $1s^{-1}3s\ ^1S_0$ state in the presence of N_L+1 laser photons. Coupling between the two degenerate atom-photon product states leads to symmetric splitting of the energies of the two resulting laser-dressed states with respect to the undressed resonance energy. The condition that must be satisfied for this so-called Autler-Townes splitting to be observable is that the Rabi frequency [10] associated with the laser coupling between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ must be greater than the Auger decay width of the core-excited states. The minimum laser intensity required to observe the Autler-Townes doublet—and, thus, the suppression of absorption at the laser-unperturbed resonance energy—is of the order of 10^{12} W/cm^2 [7]. This intensity is so high that the few-level models normally used in quantum optics [10] are unsuitable. For a detailed description of the numerical approach adopted in Ref. [7], please see Ref. [9].

The magnitude of the laser-induced suppression of the x-ray absorption cross section at the $1s \rightarrow 3p$ resonance depends on the value of the energy splitting between the $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ states. The suppression of x-ray absorption is observable only if the true energy splitting does not differ from the dressing-laser photon energy (1.55 eV) by much more than the effective width of the $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ states. The situation is illustrated in Fig. 1. In the following, we employ two different theoretical approaches to calculate an improved value for the energy splitting between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$. We calculate the resulting x-ray absorption near-edge structure of laser-dressed Ne and find good agreement with the predictions of Ref. [7].

II. ENERGY SPLITTING BETWEEN $1s^{-1}3s\ ^1S_0$ AND $1s^{-1}3p\ ^1P_1$

Our first method for determining an improved value for the energy splitting between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ is based on the exact nonrelativistic Hamiltonian [11], which may be partitioned as follows:

$$\hat{H} = \hat{H}_0 + \hat{V}^{(2)} - \hat{V}^{(1)}, \tag{1}$$

where, choosing a diagonal representation,

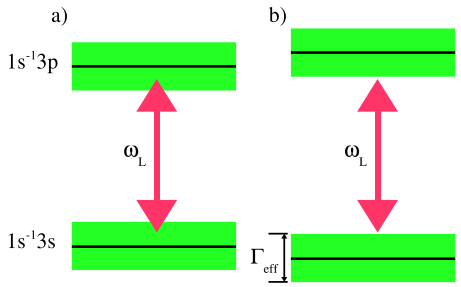

FIG. 1. (Color online) The laser photon energy ω_L is kept fixed. (a) One possibility is that the true energy splitting between the $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ states agrees with ω_L to within the decay width Γ_{eff}. (b) Another possibility is that the transition between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ is not in resonance with ω_L.

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$$\hat{H}_0 = \sum_p \left\langle \varphi_p \left| -\frac{1}{2}\nabla^2 + V^{(\mathrm{HS})}(r) \right| \varphi_p \right\rangle \hat{c}_p^\dagger \hat{c}_p = \sum_p \varepsilon_p \hat{c}_p^\dagger \hat{c}_p, \tag{2}$$

is the mean-field Hamiltonian based on the Hartree-Slater potential $V^{(\mathrm{HS})}(r)$ calculated using Ref. [8],

$$\hat{V}^{(2)} = \frac{1}{2}\sum_{pqrs} v_{pqrs}\hat{c}_p^\dagger \hat{c}_q^\dagger \hat{c}_s \hat{c}_r \tag{3}$$

is the Coulomb two-body operator (v_{pqrs} is a standard electron-electron Coulomb matrix element), and

$$\hat{V}^{(1)} = \sum_{pq} \left\langle \varphi_p \left| V^{(\mathrm{HS})}(r) + \frac{Z}{r} \right| \varphi_q \right\rangle \hat{c}_p^\dagger \hat{c}_q \tag{4}$$

represents the interaction with the residual electronic screening potential (Z is the nuclear charge). The eigenstates $|\varphi_p\rangle$ of $\hat{H}_0$ in the one-electron Hilbert space are spin orbitals forming a complete orthonormal set. The eigenvalues ε_p are orbital energies.

We use spin-singlet particle-hole excitations with respect to the Hartree-Slater ground state of the atom as zeroth-order wave functions. Thus, the energy splitting, correct through first order with respect to the perturbation $\hat{V}^{(2)}-\hat{V}^{(1)}$, is found to be $\langle 1s^{-1}3p|\hat{H}|1s^{-1}3p\rangle - \langle 1s^{-1}3s|\hat{H}|1s^{-1}3s\rangle = 1.88$ eV. This is 0.19 eV larger than the zeroth-order value (1.69 eV) used in Ref. [7].

A second, more sophisticated approach is to perform separate relativistic configuration interaction (RCI) calculations [12] for the initial state and the final state. In this approach, the radial functions of the occupied subshells in the $1s^{-1}3p\ ^1P_1$ state are numerical solutions of the Dirac-Fock equation using Desclaux's code [13]. A similar calculation for the $1s^{-1}3s\ ^1S_0$ state failed due to the indeterminacy of the off-diagonal Lagrange multiplier $\varepsilon_{1s,3s}$. To overcome this, the $1s$, $2s$, and $2p$ radial functions were taken from $1s2s^22p^63p\ ^3P_1$, and the $3s$ radial function from a $1s2s^22p^63s\ J=0$ calculation, in which $\varepsilon_{1s,3s}$ was set to 0. This $3s$ radial function was reorthogonalized to $1s$ and $2s$ in the RCI calculation. These radial functions are then corrected during the RCI process, by including single symmetry preserving replacements (e.g., $3s \rightarrow vs$, where v denotes a virtual orbital). This approach yields the same Dirac-Fock energy for Ne I $1s2s^22p^63s\ J=1$ (to 0.0001 a.u.) as obtained from a newer version of Desclaux's program [14]. At the Dirac-Fock level, the energy splitting between $1s^{-1}3p\ ^1P_1$ and $1s^{-1}3s\ ^1S_0$ was thus found to be 1.765 eV.

The RCI basis set consists of reference functions from the configuration under study and the correlation functions from configurations that are one- or two-electron replacements of the reference configuration. Subshells that are not occupied in the reference configuration are represented with virtual orbitals, whose radial functions are relativistic screened hydrogenic functions with one adjustable parameter, the effective charge Z^*. The values of the Z^*'s are determined during the energy variational process. In the calculation for Ne I, five virtual orbitals were introduced for $l \leqslant 2$ (s,p,d), three virtual orbitals for $l=3$ (f), and two virtual orbitals for $l=4$ (g). The truncation in the number of virtual orbitals was made when a new set of virtual orbitals adds no more than 1.0% to the total correlation energy in each state, resulting in a change of 1.5% in the transition energy.

TABLE I. The differentially large (>0.005 eV) correlation replacements and their correlation energies (in eV) in Ne I $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$. In this table, v denotes a virtual orbital, while x denotes either a virtual orbital or a Dirac-Fock orbital.

Replacement	$\Delta E(1s^{-1}3s\ ^1S_0)$	$\Delta E(1s^{-1}3p\ ^1P_1)$
$2p \rightarrow xp$	−0.419	−0.402
$2p3s \rightarrow vsvp$	−0.030	NA[a]
$\rightarrow vpvd$	−0.088	NA
$\rightarrow vdvf$	−0.008	NA
$2p3p \rightarrow vp^2$	NA	−0.009
$\rightarrow vd^2$	NA	−0.018
$\rightarrow vsvd$	NA	−0.013
$2p^2 \rightarrow xp^2$	−2.166	−2.151
$2s \rightarrow xs$	−0.273	−0.282
$2s3s \rightarrow vp^2$	−0.006	NA
$2s2p \rightarrow xsxp$	−1.027	−1.037
$\rightarrow xpxd$	−0.406	−0.391
Subtotal	−4.423	−4.303

[a]Not applicable (NA).

Since it is the energy splitting between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ that is of interest, only those replacements which have a different correlation effect between the two states are included in the basis set. The differentially large ones that have a net contribution of more than 5 meV to the transition energy are listed in Table I. The effect of the Breit operator was found to be small. The final result for the transition energy is 1.875 eV with an estimated accuracy of 0.050 eV. This RCI result agrees with our perturbative estimate given above.

Using a spin-dependent localized Hartree-Fock density-functional approach, Zhou and Chu obtained an energy splitting between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ of 1.84 eV [15]. A configuration-interaction calculation by Schröter *et al.* gave 1.81 eV [16]. Experimental values for the energy splitting between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$, measured by electron energy-loss spectroscopy, are 1.78(4) eV [16] and 1.95(18) eV [17].

A laser photon energy of 1.55 eV plus an Auger decay width of 0.27 eV [18] equals an energy of 1.82 eV. Hence, the available data for the energy splitting between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ suggest that if Γ_{eff} in Fig. 1 is interpreted as the Auger width, the laser is just barely in resonance with the transition between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$.

III. IMPACT ON X-RAY ABSORPTION SPECTRUM

In order to calculate the x-ray absorption near-edge structure of laser-dressed neon, we employ the computer program package FELLA [19]. The same program package was used for the calculations published in Ref. [7]. However, for the

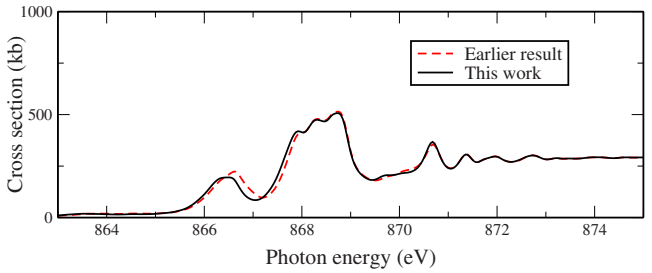

FIG. 2. (Color online) Calculated x-ray absorption cross section of atomic neon, near the Ne K edge, in the presence of an 800-nm laser field at an intensity of 10^{13} W/cm^2. The laser and x-ray polarizations are assumed to be linear and parallel to one another. The data indicated by the label "earlier result" were calculated using the input data underlying Ref. [7]. In the present work, the energy splitting between the $3s$ and $3p$ orbitals was adjusted to coincide with the energy splitting between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ calculated in Sec. II.

calculations presented here, we lower the energy of the $3s$ orbital so that the energy splitting between the $3s$ and $3p$ orbitals equals 1.875 eV (see Sec. II). All other parameters are identical to the ones listed in Ref. [7]. Note that the procedure adopted here leaves the laser-unperturbed x-ray absorption near-edge structure of Ne unchanged in comparison to Ref. [7]. Strong laser-induced suppression of resonant x-ray absorption via Autler-Townes splitting is found only when the laser and x-ray polarizations are parallel [7]. (Linear polarization is assumed.) This is the case we will focus on in the following.

Figure 2 shows the calculated x-ray absorption near-edge structure of laser-dressed neon at a laser wavelength of 800 nm and an intensity of 10^{13} W/cm^2. We compare the results calculated in Ref. [7] with our new calculation based on a more accurate value for the energy splitting between the $3s$ and $3p$ orbitals. As demonstrated in Fig. 2, the slightly increased energy splitting between the $3s$ and $3p$ orbitals, corresponding to the energy splitting between the $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ states, has almost no effect on the laser-induced suppression of resonant x-ray absorption at the $1s \rightarrow 3p$ resonance. This is in spite of the fact that, as mentioned earlier, the laser is just barely in resonance anymore with the transition between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$—provided that the width of the resonance window (Γ_{eff} in Fig. 1) is assumed to be given by the Auger width. The reason why the impact of laser dressing on the resonant x-ray absorption cross section remains strong is the following: At the required laser intensities of 10^{12} W/cm^2 or higher (see Sec. I), the core-excited states are not only broadened by Auger decay but also by laser-driven ionization of the Rydberg electron ($3s$ or $3p$). This effectively increases the width of the resonance window for laser dressing. At 10^{13} W/cm^2, the resonance window is more than 0.5 eV wide, i.e., laser ionization of the Rydberg electron occurs at a rate that is comparable to the Auger decay rate [7]. Therefore, the laser remains in approximate resonance with the transition between the $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ states.

In order to illustrate more clearly how the dressing laser affects the x-ray absorption near-edge structure of atomic

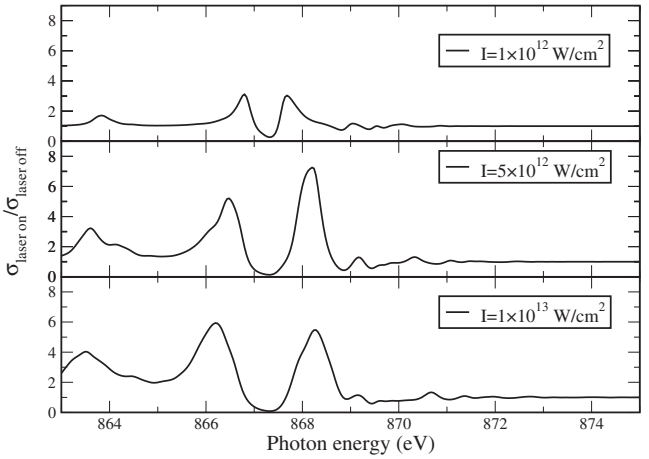

FIG. 3. The ratio between the calculated laser-on and laser-off x-ray absorption cross sections is depicted for three different laser intensities. The laser wavelength assumed is 800 nm. Parallel laser and x-ray polarizations are assumed.

neon, we plot in Fig. 3 the ratio between the laser-on and laser-off x-ray absorption cross sections for three different laser intensities. Above an x-ray photon energy of about 872 eV, this ratio is approximately equal to 1, i.e., in this x-ray energy regime, the laser has little impact on the x-ray absorption cross section. For all three laser intensities shown in Fig. 3, the x-ray absorption cross section at the energy of the laser-unperturbed $1s \rightarrow 3p$ resonance is noticeably suppressed. A laser intensity of 10^{13} W/cm^2 is found to make neon practically transparent at the resonant x-ray photon energy.

Since the laser-induced transparency is a consequence of the formation of an Autler-Townes doublet, there are also regions where the x-ray absorption cross section of laser-dressed Ne is enhanced, rather than suppressed, relative to the laser-off case. For instance, at a laser intensity of 5×10^{12} W/cm^2, the x-ray absorption cross section right above a photon energy of 868 eV is enhanced by more than a factor of 7. In this sense, one can use high-intensity laser dressing not only to make a normally strongly absorbing gas transparent to x rays, but also to make an otherwise transparent gas a more efficient absorber for x rays.

Another feature we would like to point out is the appearance of a peak below 864 eV. This peak is a little more than 1.55 eV below the energy needed to excite Ne from the atomic ground state to the $1s^{-1}3s\ ^1S_0$ state. The peak shifts to lower x-ray photon energy and is broadened as the laser intensity increases. In the limit of low laser intensity, this peak would be interpreted as a signature of the laser-assisted two-photon (one laser photon plus one x-ray photon) transition from the atomic ground state to the $1s^{-1}3s\ ^1S_0$ state. Note that this feature appears at an x-ray photon energy where the laser-off cross section is very small.

IV. CONCLUSIONS

According to our RCI calculation, the energy splitting between the $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ states of atomic neon is

1.875(50) eV. An estimate obtained using first-order nonrelativistic many-body perturbation theory gives a value of 1.88 eV. Even though this is larger by 0.19 eV than the energy splitting between $1s^{-1}3s\ ^1S_0$ and $1s^{-1}3p\ ^1P_1$ assumed in Ref. [7], we find strong laser-induced suppression of x-ray absorption at the $1s \rightarrow 3p$ resonance at a dressing-laser intensity of 10^{12} W/cm^2 or higher. This effect is assisted by the fact that the intense laser field shortens the lifetime of the core-excited states and thus broadens the resonance window available for the formation of an Autler-Townes doublet. We also find that at certain x-ray photon energies, laser dressing of neon may be employed to significantly enhance x-ray absorption. In summary, the theoretical studies presented here lend support to the prediction [7] that high-intensity laser dressing at a wavelength of 800 nm may be used to substantially modify, and thus control, the x-ray absorption near-edge structure of Ne near the $1s$ threshold.

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[1] A. Kivimäki, S. Heinäsmäki, M. Jurvansuu, S. Alitalo, E. Nõmmiste, H. Aksela, and S. Aksela, J. Electron Spectrosc. Relat. Phenom. **114–116**, 49 (2001).
[2] Y. Shimizu, H. Yoshida, K. Okada, Y. Muramatsu, N. Saito, H. Ohashi, Y. Tamenori, S. Fritzsche, N. M. Kabachnik, H. Tanaka, and K. Ueda, J. Phys. B **33**, L685 (2000).
[3] M. Coreno, L. Avaldi, R. Camilloni, K. C. Prince, M. de Simone, J. Karvonen, R. Colle, and S. Simonucci, Phys. Rev. A **59**, 2494 (1999).
[4] T. Hayaishi, E. Murakami, Y. Morioka, E. Shigemasa, A. Yagishita, and F. Koike, J. Phys. B **28**, 1411 (1995).
[5] H. Aksela, S. Aksela, J. Tulkki, T. Åberg, G. M. Bancroft, and K. H. Tan, Phys. Rev. A **39**, 3401 (1989).
[6] T. W. Gorczyca, Phys. Rev. A **61**, 024702 (2000).
[7] C. Buth, R. Santra, and L. Young, Phys. Rev. Lett. **98**, 253001 (2007).
[8] F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice-Hall, Englewood Cliffs, NJ, 1963).
[9] C. Buth and R. Santra, Phys. Rev. A **75**, 033412 (2007).
[10] P. Meystre and M. Sargent III, *Elements of Quantum Optics* (Springer, Berlin, 1991).
[11] A. L. Fetter and J. D. Walecka, *Quantum Theory of Many-Particle Systems* (McGraw-Hill, Boston, 1971).
[12] L. Pan, D. R. Beck, and S. M. O'Malley, J. Phys. B **38**, 3721 (2005).
[13] J. P. Desclaux, Comput. Phys. Commun. **9**, 31 (1975).
[14] J. P. Desclaux, in *Methods and Techniques in Computational Chemistry*, edited by E. Clementi (STEF, Cagliari, Italy, 1993), Vol. A, p. 253.
[15] Z. Zhou and Shih-I Chu, Phys. Rev. A **75**, 014501 (2007).
[16] C. D. Schröter, L. Avaldi, R. Camilloni, G. Stefani, M. Zitnik, and M. Stuhec, J. Phys. B **32**, 171 (1999).
[17] A. P. Hitchcock and C. E. Brion, J. Phys. B **13**, 3269 (1980).
[18] V. Schmidt, *Electron Spectrometry of Atoms using Synchrotron Radiation* (Cambridge University Press, Cambridge, 1997).
[19] C. Buth and R. Santra, FELLA, Argonne National Laboratory, Argonne, Illinois, USA, 2008, Version 1.3.0, with contributions by Mark Baertschy, Kevin Christ, Chris H. Greene, Hans-Dieter Meyer, and Thomas Sommerfeld.