

Effect of Electron Capture on Spectral Line Broadening in Hot Dense Plasmas

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(Received 3 October 2019; revised manuscript received 10 December 2019;
accepted 24 December 2019; published 5 February 2020)

Accurate calculation of spectral line broadening is important for many hot, dense plasma applications. However, calculated line widths have significantly underestimated measured widths for $\Delta n = 0$ lines of Li-like ions, which is known as the isolated-line problem. In this Letter, scrutinization of the line-width derivation reveals that the commonly used expression neglects a potentially important contribution from electron-capture. Line-width calculations including this process are performed with two independent codes, both of which removed the discrepancies at temperatures below 10 eV. The revised calculations also suggest the remaining discrepancy scales more strongly with electron temperature than the atomic number as was previously suggested.

DOI: [10.1103/PhysRevLett.124.055003](https://doi.org/10.1103/PhysRevLett.124.055003)

Spectral lines observed from plasmas are shifted and broadened due to the atomic wave functions being perturbed by nearby electrons and ions. While accurate understanding of line broadening is crucial for a number of applications, including plasma diagnostics [1–4] and opacity calculations [5–9], persistent disagreement between measurements and models indicates that line-broadening theory is not sufficiently accurate [10,11].

One of these outstanding disagreements is known as the *isolated-line* problem [12–14]. This problem centers around the $1s^2 2p-1s^2 2s$ and $1s^2 3p-1s^2 3s$ transitions of Li-like ions (B III, C IV, N V, O VI, F VII, and Ne VIII), where lines are broadened mostly due to atomic-states being perturbed by plasma electrons (via electron-impact collisions) [14]. In the 1990s, multiple high-fidelity experiments [15–19] were performed to measure the widths of these transitions with independent temperature and density diagnostics. The experiments found that calculations [20–22] underpredicted the measured widths. The discrepancy is most severe for the B III $1s^2 2p-1s^2 2s$ and the Ne VIII $1s^2 3p-1s^2 3s$ lines, where the calculated widths are half of the measured values.

Since this discrepancy is observed over multiple platforms, the experiments are thought to be correct. The Glenzer [15–17] experiment was a gas-liner pinch discharge and the Belgrade experiment [19] a linear arc discharge. Both experiments used a light element (hydrogen and helium, respectively) as the driver gas, and the elements of interest are tracers. Both sets of experiments used optical Thomson scattering to derive electron densities and temperatures. Thus, the model-data line-width discrepancies

consistently observed from these experiments raised a question on the line-broadening model accuracy.

The observed discrepancy is particularly puzzling because the mechanism for electron broadening was considered well understood and the key calculations were experimentally validated. Baranger [23] derived that line widths can be directly calculated from electron-collision cross sections. However, as shown in Griem *et al.* [12], line widths calculated with cross sections from state-of-the-art scattering models [24] (which have been experimentally verified [25]) disagree with measured line widths. Interestingly, semiclassical calculations somehow showed better agreement, but it was found to be coincidence due to improper treatment of penetrating collisions [24,26]. This long-standing isolated line problem has been a challenge for line-broadening community and raised an important question: why do validated collision calculations fail to reproduce the widths of these lines?

There have been several attempts to explain this discrepancy since the 1990s, but a resolution has yet to be found. Griem *et al.* [12] speculated that unnoticed turbulence in the plasma may be adding extra broadening to the lines. This hypothesis was refuted by Alexiou *et al.* [27], showing that the data contained no signs of turbulence. Iglesias [28] suggested that the discrepancy could be resolved if the atomic populations were out of local thermodynamic equilibrium, but Griem and Ralchenko [29] showed that the populations are very close to equilibrium. Further studies at the Spectral Line Shapes in Plasmas Code-Comparison Workshops found no new insight into the problem [14,30,31]. Interestingly, Duan *et al.* [32] showed excellent agreement for the B III $1s^2 2p-1s^2 2s$ line width at the one of

the workshops. However, they could not provide physical reasoning for better agreement and ended their paper by emphasizing the necessity of further scrutiny.

In this Letter, we provide a resolution and physical explanation for the B III $1s^22p-1s^22s$ discrepancy as well as a partial resolution for the $1s^23p-1s^23s$ discrepancies. We revisit Baranger's [23] derivation for the line width formula, reiterating that line-width calculations are incomplete unless all possible electron-impact collision channels are included. Generally, only elastic and inelastic collisions have been included in the broadening calculations, but electron-capture (hereafter EC) processes have never been included. By performing complete line-width calculations with the EC processes included, the widths of all Li-like ions increased. Some of the widths increased significantly; for example, the B III $1s^22p-1s^22s$ width now agrees with experiment (a result obtained by two independent codes). The increase in the widths of the $1s^23p-1s^23s$ lines resolves the discrepancies below 10 eV. Above 10 eV, there are still discrepancies which trend strongly with temperature and are not dependent on the atomic species.

For an isolated line, the line shape, $I(\omega)$, is well approximated as a Lorentzian,

$$I(\omega) \propto \frac{1}{(\omega - \omega_0^a - \mathcal{H}_R)^2 + \mathcal{H}_I^2} \quad (1)$$

where ω is the photon frequency; ω_0^a is the unperturbed atomic-transition frequency; and \mathcal{H}_R , \mathcal{H}_I are the shift and width, respectively, due to electron perturbations. The width is approximated using T matrices (or collision amplitudes) as follows (atomic units used throughout) [23],

$$\begin{aligned} \mathcal{H}_I = n_e \lambda_T^3 \int_0^\infty d^3 \mathbf{k} e^{-\beta \frac{1}{2} k^2} & \left[\Im \langle u\mathbf{k} | T | u\mathbf{k} \rangle - \Im \langle l\mathbf{k} | T^* | l\mathbf{k} \rangle \right. \\ & \left. + 2\pi \int_0^\infty d^3 \mathbf{k}' \langle u\mathbf{k} | T | u\mathbf{k}' \rangle \langle l\mathbf{k}' | T^* | l\mathbf{k} \rangle \delta(E_{\mathbf{k}} - E_{\mathbf{k}'}) \right], \end{aligned} \quad (2)$$

where n_e is the electron density, λ_T is the thermal de Broglie wavelength, β is the inverse temperature, u and l denote the upper and lower states (respectively) of the atom, and \mathbf{k} and \mathbf{k}' are states of the perturbing plasma electron. The three terms in the square brackets are considered to be the broadening of the upper state, lower state, and the interference between them [20].

The upper- and lower-state broadening terms can be alternatively defined as a function of the total collision cross section using the optical theorem [20,23], i.e., (shown for the lower broadening term):

$$\Im \langle l\mathbf{k} | T | l\mathbf{k} \rangle = -\frac{|\mathbf{k}|}{2} \sum_{l'} \hat{\sigma}_{l \rightarrow l'}(\mathbf{k}), \quad (3)$$

where $\hat{\sigma}_{l \rightarrow l'}(\mathbf{k})$ is the total cross section from state l to l' with initial perturber state \mathbf{k} . Here, Baranger provided physical insight into the electron broadening problem: the ensemble of random electron perturbations on line widths is expressed as the probability-weighted average [i.e., $\lambda_T^3 \exp(-\beta k^2/2)$] over all possible electron-collision cross sections between unperturbed states. Since the integration of Eq. (2) results in an electron collision rate, this expression suggests (or indicates) that electron broadening can be interpreted as a natural broadening induced by electron-collision processes [33].

While this insight is ingenious, Eq. (3) is slightly misleading because it obscures the necessity of electron-capture processes. To make this point clearer, we share an abridged version of the derivation below. To connect the lower-broadening term to the cross sections, the following property of the T matrix is used:

$$\Im \langle l\mathbf{k} | T | l\mathbf{k} \rangle = -\pi \langle l\mathbf{k} | T^\dagger \delta(E - H_0) T | l\mathbf{k} \rangle, \quad (4)$$

where E and H_0 are the energy and noninteracting Hamiltonian of the atom plus perturber system. To evaluate the product of operators, we use the resolution of identity,

$$1 = \sum_{l'\mathbf{k}'} |l'\mathbf{k}'\rangle \langle l'\mathbf{k}'|, \quad (5)$$

so that

$$\begin{aligned} \Im \langle l\mathbf{k} | T | l\mathbf{k} \rangle = -\pi \sum_{l'\mathbf{k}'} \langle l\mathbf{k} | T^\dagger | l'\mathbf{k}' \rangle \langle l'\mathbf{k}' | T | l\mathbf{k} \rangle \\ \times \delta(E_{l\mathbf{k}} - E_{l'\mathbf{k}'}). \end{aligned} \quad (6)$$

Since $\int_{\mathbf{k}} |\langle l\mathbf{k} | T | l'\mathbf{k}' \rangle|^2$ is proportional to a cross section [34,35], one can rewrite Eq. (6) as a function of cross sections.

However, we have to be cautious when converting Eq. (6) to Eq. (3). It is imperative to note that the identity resolution, Eq. (5), is satisfied only when the summation goes over all possible states, i.e., including both bound and free (or ionized) states for the atomic state l' and for the perturbing electron state \mathbf{k}' . In other words, one must include the following processes in the summation: collisional excitation or de-excitation [Figs. 1(a) and 1(b) where $l' = \text{bound}$, $\mathbf{k}' = \text{free}$], collisional ionization [Fig. 1(c) where

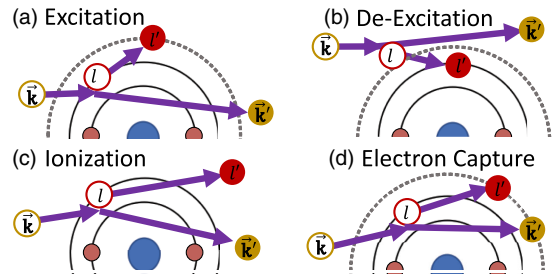


FIG. 1. Cartoon of possible binary-collision channels: (a) excitation, (b) de-excitation, (c) ionization, and (d) recombination or electron capture.

$l' = \text{ionized}, \mathbf{k}' = \text{free}$], and electron capture [Fig. 1(d) where $l' = \text{bound}, \mathbf{k}' = \text{bound}$]. The importance of collisional ionization was investigated and found to be negligible [36]; however, electron-capture has not been investigated.

To illustrate how the electron-capture process differs from the other processes, we pick out the contribution for the case of $l = 1s^22s$ and $l' = 1s^22p$ from Eq. (6), which is this:

$$\sum_{\mathbf{k}'} |\langle 2s\mathbf{k} | T | 2p\mathbf{k}' \rangle|^2 \delta(E_{2s\mathbf{k}} - E_{2p\mathbf{k}'}) = \int d\mathbf{k}' |\langle 2s\mathbf{k} | T | 2p\mathbf{k}' \rangle|^2 \delta(E_{2s} + E_{\mathbf{k}} - E_{2p} - E_{\mathbf{k}'}) + \sum_{n'l'} |\langle 2s\mathbf{k} | T | 2pn'l' \rangle|^2 \delta(E_{2s} + E_{\mathbf{k}} - E_{2p} - E_{n'l'}), \quad (7)$$

(note: $1s^2$ is common to all atomic states and omitted in the state label for brevity). For the right-hand side, we write out the integral over \mathbf{k}' and sum over \mathbf{k}' (which turns into a set of nl states). The first term with the integral is the contribution from collisional excitation of $2s$ to $2p$ where the energy difference is 6.0066 eV [37]. The delta function ensures conservation of energy between the initial and final states. Thus, through this process, the incident electron (i.e., perturbing electron) collides and excites the atom from $2s$ to $2p$ and leaves the system with 6 eV less energy than it started, $E_{\mathbf{k}'} = E_{\mathbf{k}} - 6$ eV. Therefore, as long as the incident-electron energy, $E_{\mathbf{k}}$, is greater than the atomic excitation energy, this collisional excitation happens, which can be seen from the nonzero cross section above $E_{\mathbf{k}} > 6$ eV in Fig. 2.

The second term of Eq. (7) is the contribution from EC. This term suggests that there is a nonzero contribution to broadening even if $E_{\mathbf{k}}$ is less than 6 eV. In order for this to happen, the scattered electron has to go to a negative-energy (bound) state, becoming trapped by the atomic potential. Since the bound states ($n'l'$) have discrete energies, the process is only allowed when $E_{\mathbf{k}} = E_{n'l'} + 6$ eV. This EC contribution is shown as

discrete contributions below 6 eV in Fig. 2. It is also worth noting that this EC contribution on line widths becomes more important at lower temperature due to the larger abundance of such low-energy (< 6 eV) free electrons. This is indicated by the weighting function, $\exp(-\beta k^2/2)$ of Eq. (2) and the gray curves in Fig. 2.

This EC contribution is easily overlooked when the line width is evaluated as a sum over cross sections [like in Eq. (3)] because $\hat{\sigma}_{l \rightarrow l'}(\mathbf{k})$ is the cross section between atomic states l and l' , omitting the cross section from a Li-like state (e.g., $l = 1s^22s$) to a doubly excited Be-like state (e.g., $1s^22p3p$ where $l' = 1s^22p$ and $\mathbf{k}' = 3p$). Equation (3) is still correct as long as l' is extended to include recombination to lower-charge states, but to our knowledge, it has never been explicitly stated in the literature. Below, we evaluate how important EC is for the longstanding isolated line discrepancy.

We first evaluate the impact of EC by using a Coulomb-Born-Exchange (CBE) calculation [38]. CBE uses the approximation

$$\Im \langle l\mathbf{k} | T | l\mathbf{k} \rangle \approx -\pi \langle l\mathbf{k} | V \delta(E - H_0) V | l\mathbf{k} \rangle, \quad (8)$$

where V is a Coulomb interaction that includes exchange (i.e., accounting for indistinguishability of electrons). The effect of EC in this approximation is only going to affect the broadening of the upper and lower states and will not contribute to the interference term. This is due to the delta function in Eq. (2), which means that the set of intermediate states will have the same energy as the starting state.

When line widths are computed without the EC contribution [first term of Eq. (7) alone], our CBE calculation reproduces the familiar factor-of-two underestimate ($w_{\text{calc}}/w_{\text{meas}} = 0.55$), as expected. By including the EC contribution [i.e., both terms of Eq. (7)], the line width increases and successfully reproduces the measured line width, $w_{\text{calc}}/w_{\text{meas}} = 1.02$, within the measurement error (Table I).

This result implies that the line-width disagreement was caused by the lack of EC contribution; however, it was not immediately clear why Griem *et al.* [12] could not reproduce the measured width using convergent-close-coupling

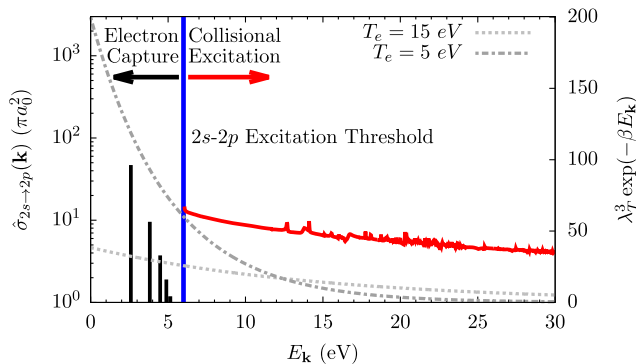


FIG. 2. Shown is the cross section for different values of the incident-electron energy, $E_{\mathbf{k}}$. Excitation ($E_{\mathbf{k}} > 6$ eV) is shown in red, while electron capture ($E_{\mathbf{k}} < 6$ eV) is shown as the discrete black features below the excitation threshold. Boltzmann distributions for $T_e = 15$ eV (dotted line) and $T_e = 5$ eV (dash-dotted line) are also shown to demonstrate that electron capture becomes more important at lower temperatures.

TABLE I. CBE and CCC widths for B III $2s-2p$ line at $T_e = 10.6$ eV and $n_e = 1.81 \times 10^{18}$ e/cm³; the measured width is 0.22 ± 0.03 Å [17]. All widths are given in Å.

	No EC	With EC
CBE	0.122	0.224
CCC	0.104 [12]	0.227

(CCC) [39] or *R*-matrix [40] methods. Both solve the Lippmann-Schwinger [34] equations exactly instead of using the approximate form [Eq. (8)]. Griem *et al.* [12] also stated that “electron impact ionization and recombination can be safely neglected.” If “recombination” refers to EC, then this statement contradicts our finding.

Our hypothesis is that Griem *et al.* [12] did not include EC when performing the sum of cross sections [Eq. (1) in [12]] and was referring to three-body recombination when stating “recombination.” Griem *et al.* [12] uses Eq. (3) for line-width calculations. While this expression requires deliberate inclusion of all processes including EC, Ref. [12] did not mention this process, and CCC does not output the EC cross sections. It is highly possible that EC contribution was overlooked in their calculations.

We can test this hypothesis by recalculating the line width using CCC that includes EC. Since CCC does not output EC cross sections, we use the more fundamental form, Eq (2), using the three *T*-matrix terms that are directly computed with CCC. This is the equation before introducing the identity resolution [Eq. (5)], and the EC effect is naturally included in the free-free *T* matrices. With this alternative way to include EC broadening, CCC also reproduced the experimental widths within 3% ($w_{\text{calc}}/w_{\text{meas}} = 1.03$). This strongly supports our hypothesis that Griem neglected EC and asserts the importance of EC on Li-like line widths. These $2s-2p$ CCC results are shown in Table I and Fig. 3(a).

These findings also seem to explain why the calculations of Duan *et al.* [32] agreed with experiments. Duan *et al.* [32] used an alternate form of Eq. (2) with *S* matrices [see Eqs. (1) and (2) in Ref. [41]], and *S* matrices are computed by taking into account the interaction of bound and free electrons using an *R*-matrix code [DARC [42]], which has the same physics as CCC [43]. As a result, their line-width calculations naturally included the EC contributions and agreed with the measured B III $2s-2p$ widths.

In Fig. 3(a), we also display the $3s-3p$ line widths using CCC, with and without EC, for the conditions and elements from Refs. [15,16,19]. CCC without EC are taken from Ralchenko *et al.* [13]. The extra broadening due to EC is not as large as that for the $2s-2p$ line. This behavior is expected because as the principal quantum number increases, the number of available EC channels become smaller and the interaction between atomic and perturbing electrons becomes weaker.

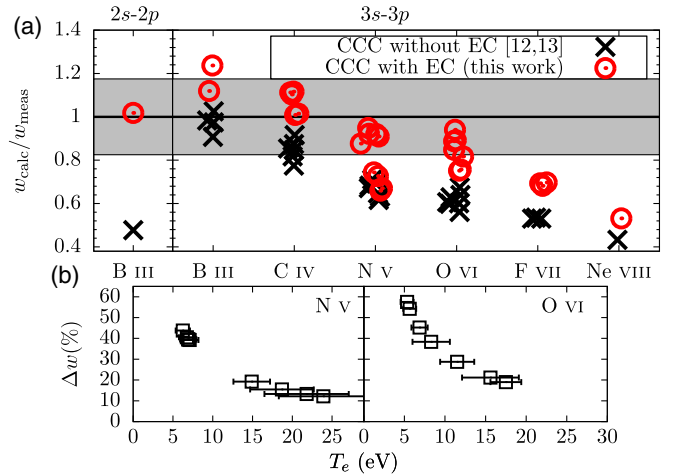


FIG. 3. (a) Ratios of calculated width over measured width by CCC without EC (black “x”; Ref. [12,13]) and CCC with EC (red circle). Each individual point corresponds to a different temperature and density. The spread in the red circles comes from T_e dependence on EC contribution. (b) The percent increase in the width due to EC on N v and O vi as a function of T_e . The grey bands indicate the average level of measurement uncertainty across all measurements.

It is interesting to note that some calculations show bigger EC corrections than others within the same element. For example, for N V lines, some calculations show notable changes due to EC ($\sim 60\%$) while others do not ($\sim 10\%$). This is caused by strong dependence of EC on electron temperature, T_e . To clarify the temperature dependence on the EC effect, we replotted N v and O vi results in Fig. 3(b); their line-width increases due to EC is plotted as a function of T_e . This figure confirms the strong correlation between EC broadening and T_e as discussed earlier.

Motivated by the temperature dependence of EC, we replotted all the elements considered in Fig. 3 as a function of T_e in Fig. 4. Different colors are assigned to different elements and the different symbols are assigned to the different experiments. We find that, for all conditions less than ~ 10 eV, the inclusion of EC is enough to remove the previously reported discrepancies with measured widths, independent of element and independent of experiments.

Figure 4 also suggests that the remaining discrepancies scale more strongly with T_e than atomic number (as originally shown by Ref. [13]). This may suggest that either the experiment has more low-energy free electrons than inferred by Thomson scattering or that there is additional temperature-dependent broadening mechanism missing from current theory.

The impact of EC on other line widths needs to be carefully scrutinized. EC cannot happen in neutral radiators because a net attractive charge is necessary to trap the incident electron into a bound state. The EC broadening becomes important for lower principal quantum number n at lower temperature. Its importance also scales with the

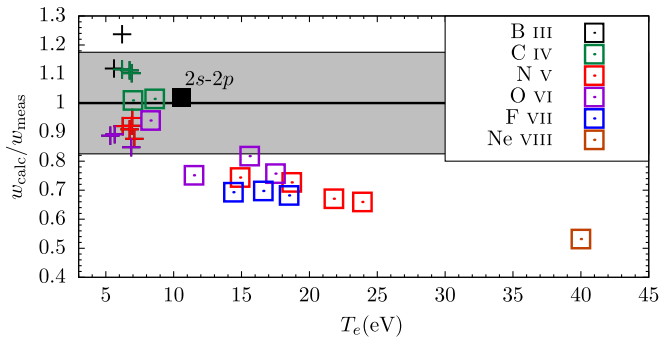


FIG. 4. Ratio of calculated to measured line widths for the $2s-2p$ transition (solid black box) and $3s-3p$ transitions (open, colored boxes for the Bochum experiment [15–18] and crosses for the Belgrade experiment [19]) as a function of temperature. Shaded grey indicates the average experimental errors across all experiments. Typical errors in T_e range from 1–3 eV, except for Ne, where the temperature errors are ~ 10 eV. The remaining discrepancy is worse with increasing temperature; data points below ~ 10 eV are within the experimental uncertainty.

number of available EC channels. One important area of investigation is its impact on K -shell spectroscopy. The density of hot plasma is often diagnosed by line shapes of H-like and He-like lines. Based on our CBE calculations, it has a significant effect on alpha lines ($n = 1-2$) but is negligible on beta and above ($n = 1-3$ or higher); this is due to the reduced number of EC channels and weaker interaction with increasing n . Negligible impact on higher n lines is good news because alpha lines are not density sensitive and are not used for density diagnostics, density is usually determined from high- n line shapes. Models of alpha lines have additional complexities due to ion dynamics and strong collision treatment, and have never been experimentally validated due to its high sensitivity to Doppler and instrumental broadening, and plasma gradients. EC broadening needs to be added to this complexity list for future investigations of alpha lines. Its impact on more complex lines needs to be investigated too. For example, recently, L -shell line widths measured at stellar interior conditions are significantly broader than calculated widths [44]. L -shell line opacities are line transitions starting from $n = 2$ (e.g., $n = 2-3$, $2-4$, etc.) and the additional broadening due to EC is expected to be strongest for the $n = 2-3$ lines. If EC is important, it could affect the accuracy of L -shell opacity calculations that many plasma simulations rely on.

We thank Doctors Y. Ralchenko, E. Stambulchik, S. Alexiou, M.-A. Schaeuble, M. H. Montgomery, J. Colgan, and C. Iglesias for their feedback and comments on the manuscript. T. A. G. is supported by the U.S. Department of Energy, Office of Science Early Career Research Program, Office of Fusion Energy Sciences under FWP-14-017426. Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and

Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy’s National Nuclear Security Administration under Contract No. DE-NA-0003525. The work of C. F., D. K., and M. Z. was carried out under the auspices of the U.S. Department of Energy by Los Alamos National Laboratory under Contract No. 89233218CNA000001. This Letter describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the Letter do not necessarily represent the views of the U.S. Department of Energy or the United States Government. D. V. F., A. S. K., and I. B. acknowledge the funding of the Australian Research Council and the resources and services from the National Computational Infrastructure and the Pawsey Supercomputer Centre, which are supported by the Australian and Western Australian Governments.

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