Calculating Opacity in Hot, Dense Matter Using Second-Order Electron-Photon and Two-Photon Transitions to Approximate Line Broadening

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Calculations of the opacity of hot, dense matter require models for plasma line broadening. However, the most general theories are too complex to calculate directly and some approximation is inevitably required. The most widely used approaches focus on the line center, where a Lorentzian shape is obtained. Here, we demonstrate that in the opposite limit, far from the line center, the opacity can be expressed in terms of second-order transitions, such as electron-photon and two-photon processes. We suggest that this insight could form the basis for a new approach to improve calculations of opacity in hot, dense matter. Preliminary calculations suggest that this approach could yield increased opacity away from absorption lines.

The opacity of hot, high density material is important for both terrestrial and astrophysical applications, for example in modeling inertial confinement fusion [1–3], stellar physics [4,5], and planetary interiors [6–8]. Predicted opacities, alongside equations of state and the abundances of heavy elements, are key inputs to solar models [9]. An increase in predicted opacities could help to reconcile discrepancies between solar models and helioseismic measurements that presently exist [10].

Some recent measurements of iron under solar conditions have yielded opacities in excess of predictions [11,12]. In particular, increased opacity was observed in regions between characteristic absorption lines. Such regions make the strongest contribution to the Rosseland mean opacity, which encapsulates the influence of the opacity on radiation transport [13]. However, the measured opacities cannot be explained within the current theoretical consensus [14,15] and are hard to reconcile with the upper limit for single-photon dipole absorption imposed by f-sum rules [16].

Such experimental findings have prompted recent interest in two-photon transitions as a possible source of opacity. Two-photon transitions are obtained from second-order perturbation theory and can be interpreted as the simultaneous absorption or emission of two photons. Since the energy required for the transition may be divided between the two photons in any ratio, two-photon processes yield a continuum of absorption reminiscent of the additional opacity seen experimentally. Furthermore, being at second order in perturbation theory, two-photon opacity may exceed the limits of f-sum rules. However, published results have reached mixed conclusions, with the most detailed calculations finding that two-photon rates should be negligible under the relevant conditions [17–19].

Two-photon processes are not the only mechanism unconstrained by the f-sum rule. In dense plasmas, the absorbing ions are also perturbed by the plasma electrons and ions. This results in broadening of the spectral lines, a process that has been widely explored [20–23]. We can, however, describe line broadening in a novel way by identifying a process that is the collisional analog of a two-photon transition. This process, which we call the electron-photon process, consists of a photon absorption and an electron collision occurring simultaneously. Although line broadening is not usually approached in this way, the appearance of some satellite lines has been explained in terms of electron-photon transitions [24,25].

Both the electron-photon description and more conventional line broadening approaches aim to describe the same physical process. However, the extent to which they are formally equivalent is not immediately apparent. A similar uncertainty exists in the case of two-photon transitions. While it has been suggested that two-photon processes might be interpreted as line broadening by background radiation fields [26], this relationship remains relatively unexplored.

In this work, we demonstrate that we can approximate line broadening in the far wings of the lines with the electron-photon cross section. The formalism developed for electron broadening is easily extended to the case of broadening by radiation. This allows us to give an expression for broadening due to photons, which can be approximated by the two-photon cross section in the line wings. This treatment suggests a new perspective on opacity that may allow improved calculations in spectral regions away from absorption lines, which are highly relevant to stellar modeling.

We begin by considering line broadening by collisions and its relationship to the second-order electron-photon process. The overall line shape is due to both the electrons and ions. It is often assumed that the ions can be treated statically, while the electrons make both a static and dynamic contribution. The line shape can then be written as [27,28]

$$I(\omega) = \int d\mathbf{F} W(\mathbf{F}) J(\omega, \mathbf{F}), \tag{1}$$

with

$$J(\omega, \mathbf{F}) = -\frac{1}{\pi} \text{Im} \sum_{a,b,c,d} \langle b|\boldsymbol{\epsilon} \cdot \mathbf{r}|a\rangle \langle c|\boldsymbol{\epsilon} \cdot \mathbf{r}|d\rangle$$
$$\times \langle ab|[\Delta\omega - \mathbf{r} \cdot \mathbf{F}/\hbar - \phi(\omega)]^{-1}\rho|cd\rangle. \tag{2}$$

Here, $\mathbf{F}$ represents the static microfield, which introduces a line shift. This becomes a line broadening due to integration over the statistical distribution, $W(\mathbf{F})$. Since this work is concerned with broadening by electron collisions, we can neglect $\mathbf{F}$ for brevity without loss of generality. The sum runs over the atomic states, both bound and continuum. The density operator ρ gives the statistical populations of these states. Notably, this expression does not necessarily reduce to a form that would be bound by the f-sum rule.

In order to make a comprehensive comparison with the second-order electron-photon process, it is necessary to start with a general form for the broadening operator $\phi(\omega)$, where the matrix elements are given by [29]

$$\phi_{ab,cd}(\omega) = \frac{1}{\hbar^2} \int \frac{d\mathbf{k}}{(2\pi)^3} \int_{-\infty}^{\infty} \frac{d\Omega}{2\pi} S(k, \Omega)$$
$$\times \left(\sum_e \delta_{bd} \frac{V_{ae}(\mathbf{k})V_{ec}(-\mathbf{k})}{\Delta\omega_{eb} - \Omega + i\eta} \rho_e \rho_c^{-1} + \sum_e \delta_{ac} \frac{V_{de}(-\mathbf{k})V_{eb}(\mathbf{k})}{\Delta\omega_{ae} - \Omega + i\eta} \right.$$
$$\left. - V_{ac}(\mathbf{k})V_{db}(-\mathbf{k}) \left[\frac{1}{\Delta\omega_{cb} - \Omega + i\eta} + \frac{\rho_a \rho_c^{-1}}{\Delta\omega_{ad} - \Omega + i\eta} \right] \right). \tag{3}$$

We define ω such that positive values correspond to absorption. The frequency detuning is then defined as $\Delta\omega_{ab} = (\omega_b - \omega_a) - \omega$. $V_{ij}(\mathbf{k}) = \langle i|V(\mathbf{k})|j\rangle$ are the matrix elements of the electron-radiator interaction [30]. The dynamic structure factor of the plasma electrons $S(k, \Omega)$ encompasses much of the physics of the collisions, which can alternatively be thought of in terms of absorption or emission of plasmons.

We first consider the case where we are far from the line center. Making an expansion of Eq. (2) in orders of $\phi/\Delta\omega$, we obtain [25,28]

$$J(\omega) = -\frac{1}{\pi} \text{Im} \sum_{a,b,c,d} \langle b|(\boldsymbol{\epsilon} \cdot \mathbf{r})|a\rangle \langle c|(\boldsymbol{\epsilon} \cdot \mathbf{r})|d\rangle$$
$$\times \langle ab|\left(\frac{1}{\Delta\omega} + \frac{1}{\Delta\omega}\phi(\omega)\frac{1}{\Delta\omega} - \mathcal{O}(\Delta\omega^{-3})\right)\rho|cd\rangle. \tag{4}$$

Introducing Eq. (3), the lowest order contribution in this expansion is then given by

$$J(\omega) \approx -\frac{1}{\pi} \sum_{a,b,c,d} \langle b|(\boldsymbol{\epsilon} \cdot \mathbf{r})|a\rangle \langle c|(\boldsymbol{\epsilon} \cdot \mathbf{r})|d\rangle \frac{\text{Im}\phi_{ab,cd}(\omega)}{\Delta\omega_{ab}\Delta\omega_{cd}}$$
$$\approx \frac{1}{\hbar^2} \int \frac{d\mathbf{k}}{(2\pi)^3} \sum_{a,b,c,d,e} \left[\right.$$
$$\frac{d_{ba}d_{cb}V_{ae}(\mathbf{k})V_{ec}(-\mathbf{k})}{\Delta\omega_{ab}\Delta\omega_{cb}} S(k, \Delta\omega_{eb})\rho_e$$
$$+ \frac{d_{bc}d_{cd}V_{de}(-\mathbf{k})V_{eb}(\mathbf{k})}{\Delta\omega_{cb}\Delta\omega_{cd}} S(k, \Delta\omega_{ce})\rho_c$$
$$- \frac{d_{ba}d_{cd}V_{ac}(\mathbf{k})V_{db}(-\mathbf{k})}{\Delta\omega_{ab}\Delta\omega_{cd}} S(k, \Delta\omega_{cb})\rho_c$$
$$\left. - \frac{d_{ba}d_{cd}V_{ac}(-\mathbf{k})V_{db}(\mathbf{k})}{\Delta\omega_{ab}\Delta\omega_{cd}} S(k, \Delta\omega_{ad})\rho_a \right], \tag{5}$$

where we have abbreviated the dipole matrix elements according to $d_{ij} = \langle i|\boldsymbol{\epsilon} \cdot \mathbf{r}|j\rangle$. We now compare this with the rate for an electron-photon transition, calculated using second-order perturbation theory [31,32]:

$$w_{e\gamma} = \frac{2\pi}{\hbar} \int \frac{d\mathbf{k}}{(2\pi)^3} S(k, \omega_e)\pi e^2 E_0^2(\omega_\gamma)$$
$$\times \left| \sum_n \frac{\langle f|\boldsymbol{\epsilon} \cdot \mathbf{r}|n\rangle \langle n|V(\mathbf{k})|i\rangle}{E_n - E_i - \hbar\omega_e} + (e \leftrightarrow \gamma) \right|^2$$
$$\times \delta(E_f - E_i - \hbar\omega_e - \hbar\omega_\gamma), \tag{6}$$

where the positions of the electron and photon are exchanged in the second term. We can use the delta function to express the collision energy in terms of the frequency detuning:

$$\omega_e = (\omega_f - \omega_i) - \omega_\gamma = \Delta\omega_{if}. \tag{7}$$

Now taking the total opacity due to electron-photon transitions, and expanding the square modulus, we can obtain a quantity corresponding to $J(\omega)$ of Eq. (5):

$$J(\omega) \approx \frac{\hbar}{2\pi^2 e^2 E_0^2(\omega_\gamma)} \sum_{i,f} w_{e\gamma} \rho_i$$

$$\approx \frac{1}{\hbar^2} \int \frac{d\mathbf{k}}{(2\pi)^3} \sum_{i,f,m,n} \left[\frac{d_{mf} d_{fn} V_{ni}(\mathbf{k}) V_{im}(-\mathbf{k})}{\Delta\omega_{mf} \Delta\omega_{nf}} \right.$$

$$+ \frac{d_{im} d_{ni} V_{mf}(-\mathbf{k}) V_{fn}(\mathbf{k})}{\Delta\omega_{im} \Delta\omega_{in}} - \frac{d_{im} d_{fn} V_{ni}(\mathbf{k}) V_{mf}(-\mathbf{k})}{\Delta\omega_{im} \Delta\omega_{nf}}$$

$$\left. - \frac{d_{mf} d_{ni} V_{im}(-\mathbf{k}) V_{fn}(\mathbf{k})}{\Delta\omega_{mf} \Delta\omega_{in}} \right] S(k, \Delta\omega_{if}) \rho_i. \qquad (8)$$

Comparing Eqs. (5) and (8), we see that, with some relabeling of summation variables, the two expressions are identical. There is therefore exact equivalence between the electron-photon process and the first term in the line shape expansion. Thus, in the far line wings the broadened transition can be expressed in terms of electron-photon transitions. Closer to the line center, it will become necessary to include further terms in the expansion. It is possible that such terms correspond to higher-order transitions involving a photon together with two or more electron collisions.

We can also consider line broadening by background radiation. The broadening operator can be expressed in terms of the Fourier transform of the electric field autocorrelation [22]. In the radiation case, we then apply the Wiener-Khinchin theorem, which relates the autocorrelation function of the fluctuating radiation field to the spectral energy density:

$$\int_{-\infty}^{\infty} dt e^{i\omega t} \langle E(\tau) E(\tau + t) \rangle = u(\omega). \qquad (9)$$

Using this relation, we can then derive a broadening operator, analogous to Eq. (3), for broadening by background radiation:

$$\phi_{ab,cd} = \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \frac{d\Omega}{2\pi} 2\pi e^2 u(\Omega)$$

$$\times \left(\sum_e \delta_{bd} \frac{d_{ae} d_{ec}}{\Delta\omega_{eb} - \Omega - i\eta} \rho_e \rho_c^{-1} + \sum_e \delta_{ac} \frac{d_{de} d_{eb}}{\Delta\omega_{ae} - \Omega - i\eta} \right.$$

$$\left. - d_{ac} d_{db} \left[\frac{1}{\Delta\omega_{cb} - \Omega - i\eta} + \frac{\rho_a \rho_c^{-1}}{\Delta\omega_{ad} - \Omega - i\eta} \right] \right). \qquad (10)$$

Substituting this into Eq. (4), just as in the collisional case, we can obtain the two-photon rate as the lowest-order contribution,

$$J(\omega) \approx \frac{1}{\hbar^2} \sum_{i,f,m,n} \left[\frac{d_{mf} d_{fn} d_{ni} d_{im}}{\Delta\omega_{mf} \Delta\omega_{nf}} + \frac{d_{im} d_{ni} d_{mf} d_{fn}}{\Delta\omega_{im} \Delta\omega_{in}} \right.$$

$$\left. - \frac{d_{im} d_{fn} d_{ni} d_{mf}}{\Delta\omega_{im} \Delta\omega_{nf}} - \frac{d_{mf} d_{ni} d_{im} d_{fn}}{\Delta\omega_{mf} \Delta\omega_{in}} \right] 2\pi e^2 u(\Delta\omega_{if}) \rho_i. \qquad (11)$$

Subsequent terms in the expansion possibly correspond to higher-order multiphoton processes. With an appropriate definition of the energy density $u(\omega)$, the expansion includes not only stimulated two-photon processes but also processes incorporating spontaneous emission.

In principle then, a sufficiently general treatment of the line broadening would already include opacity due to second-order processes. This is the case even for second-order transitions with no directly intermediate state, such as the $1s - 2s$ case. The second-order opacity here arises from the far line wings of the $1s - 2p$ (and higher np) line, which extend into the energy range between $1s$ and $2s$. Such transitions had previously been thought of as a "pure" two-photon contribution, with no equivalent in one-photon approaches [33].

In practice, however, the general broadening operators given in Eqs. (3) and (10) are computationally intractable. As a result, conventional approaches to opacity calculations depend on approximations that are valid close to the line center [34]. In the isolated line limit, which requires $\Delta\omega \rightarrow 0$ for only the line in question [35], the line shape has the familiar Lorentzian form

$$J(\omega) = -\frac{1}{\pi} \sum_{a,b} |d_{ab}|^2 \frac{\gamma_{ab}}{\Delta\omega_{ab}^2 + \gamma_{ab}^2} \rho_a. \qquad (12)$$

In this form, the opacity can be constrained using the f-sum rule. The width is given by the diagonal components of the broadening operator [21,29]

$$\gamma_{ab} = \mathrm{Im}\phi_{ab,ab}$$

$$= \left\langle \frac{n_e v}{2} \left[\sigma_a + \sigma_b + \int d\Omega |F_a(\Omega) - F_b(\Omega)|^2 \right] \right\rangle_{\mathrm{Av}}. \qquad (13)$$

Here, F are the contributions due to elastic collisions. The $\Delta\omega \rightarrow 0$ limit may be relaxed somewhat for this contribution, for example using the approach of Lee [23]. The σ are the inelastic contribution, due to collisional and radiative rates. In particular, this includes the rate of spontaneous radiative emission, which leads to what is often referred to as the "natural" width of the state. This interpretation only holds in the line center. In the line wings, the concept of natural width should give way to two-photon transitions involving spontaneous emission.

To summarize, the most general theory for line broadening is not constrained by the f-sum rule. However, the approximation commonly used in opacity calculations [Eq. (12)] preserves the f-sum rule. We can improve upon this by including off-diagonal terms of the broadening operator that are not included in Eq. (12). These off-diagonal terms can arise either from collisions with plasma electrons or from a second photon. In our treatment, we
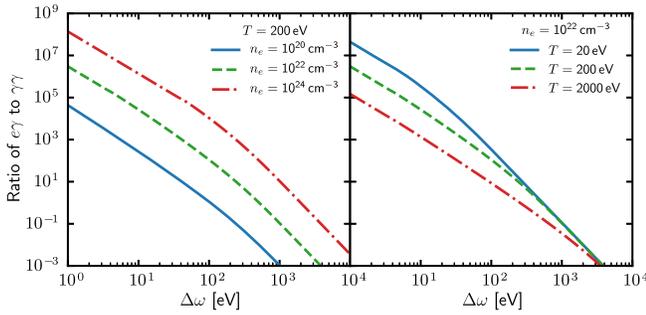
FIG. 1. The ratio of electron-photon to two-photon processes shown against detuning from the line center for typical solar conditions. The rates have been calculated in thermal equilibrium, using the RPA structure factor, which will be valid for a weakly coupled plasma [36].

FIG. 2. Calculated opacity for Ne-like iron in a plasma with $T = 180$ eV and $n_e = 1.88 \times 10^{21}$ cm^{-3}. The simplified Lorentzian broadening [Eq. (12)] is compared with an improved broadening using the electron-photon approximation [Eq. (8)]. The same atomic data have been used in both cases.

approximate these off-diagonal terms with the electron-photon and two-photon cross sections [Eqs. (8) and (11)].

Employing this approximation in the line wings opens up the possibility for a new approach to opacity calculations based on two-photon and electron-photon processes. This approach would be most accurate in the regions between lines. Since these regions contribute strongly to the Rosseland mean opacity, this new approach would be particularly suited to opacity calculations for radiative transport.

As a first step toward carrying out opacity calculations in a second-order picture, we can determine the relative importance of electron-photon and two-photon processes. If both collisions and radiation are treated in the dipole approximation, then, by comparing Eqs. (8) and (11), the ratio of the two processes can be written simply as a function of the detuning:

$$\frac{w_{e\gamma}}{w_{\gamma\gamma}} = \int \frac{d\mathbf{k}}{(2\pi)^3} k^2 \left(\frac{4\pi e^2}{k^2}\right)^2 S(k, \omega)(2\pi e^2 u(\omega))^{-1}. \quad (14)$$

This expression has been evaluated for typical solar conditions in Fig. 1. We find that the electron-photon process dominates over the two-photon process for moderate detuning ($\lesssim 100$ eV). This is consistent with calculations suggesting negligible two-photon contributions in the 6–10 Å range [19]. As a result, we focus initially on the electron-photon process. However, the two-photon process should become the more important for larger detuning, for example between the K and L shells of heavier elements.

A preliminary calculation of the opacity of neonlike iron under solar conditions is shown in Fig. 2 (see Supplemental Material for technical details [37]). The opacity calculated using the electron-photon approximation is compared with an equivalent calculation using the conventional, Lorentzian broadening. For the range shown, the opacity is dominated by L-shell bound-bound opacity. Where it is valid (i.e., in the regions between the lines), the electron-photon approach leads to an increase in opacity of up to
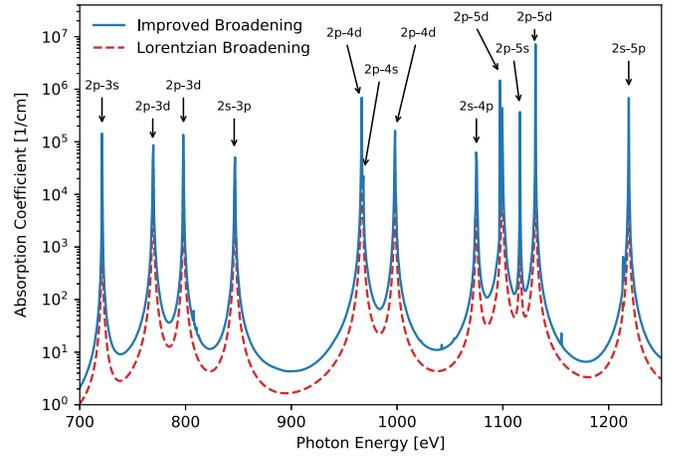
200%. Since radiation transport in hot, dense matter is dominated by these spectral regions, this suggests that the electron-photon approach could have significant impact on opacity calculations for radiation transport applications.

This result is also promising with respect to recent disagreements between theory and experiment [11]. However, the present preliminary calculation includes only a single charge state. Reaching a firm conclusion in this regard would require an electron-photon opacity calculation on a par with state-of-the-art conventional codes, incorporating a full distribution of charge states and excited states.

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[1] T. R. Dittrich, O. A. Hurricane, D. A. Callahan, E. L. Dewald, T. Döppner, D. E. Hinkel, L. F. Berzak Hopkins, S. Le Pape, T. Ma, J. L. Milovich, J. C. Moreno, P. K. Patel, H.-S. Park, B. A. Remington, J. D. Salmonson, and J. L. Kline, Phys. Rev. Lett. **112**, 055002 (2014).

[2] S. X. Hu, L. A. Collins, V. N. Goncharov, T. R. Boehly, R. Epstein, R. L. McCrory, and S. Skupsky, Phys. Rev. E **90**, 033111 (2014).

[3] E. Hill and S. Rose, High Energy Density Phys. **8**, 307 (2012).

[4] S. E. Woosley, A. Heger, and T. A. Weaver, Rev. Mod. Phys. **74**, 1015 (2002).

[5] C. Blancard, P. Cossé, and G. Faussurier, Astrophys. J. **745**, 10 (2012).

[6] D. Saumon and T. Guillot, Astrophys. J. **609**, 1170 (2004).

[7] T. Guillot, Annu. Rev. Earth Planet Sci. **33**, 493 (2005).

 [8] D. C. Swift, J. H. Eggert, D. G. Hicks, S. Hamel, K. Caspersen, E. Schwegler, G. W. Collins, N. Nettelmann, and G. J. Ackland, Astrophys. J. **744,** 59 (2012).

 [9] M. Asplund, N. Grevesse, A. J. Sauval, and P. Scott, Annu. Rev, Astron. Astrophys. **47,** 481 (2009).

[10] S. Basu and H. M. Antia, Phys. Rep. **457,** 217 (2008).

[11] J. E. Bailey *et al.*, Nature (London) **517,** 56 (2015).

[12] T. Nagayama *et al.*, Phys. Rev. Lett. **122,** 235001 (2019).

[13] W. F. Huebner and W. D. Barfield, *Opacity*, Astrophysics and Space Science Library Vol. 402 (Springer, New York, New York, NY, 2014).

[14] N. R. Badnell, M. A. Bautista, K. Butler, F. Delahaye, C. Mendoza, P. Palmeri, C. J. Zeippen, and M. J. Seaton, Mon. Not. R. Astron. Soc. **360,** 458 (2005).

[15] C. A. Iglesias and F. J. Rogers, Astrophys. J. **464,** 943 (1996).

[16] C. A. Iglesias, High Energy Density Phys. **15,** 4 (2015).

[17] R. M. More, S. B. Hansen, and T. Nagayama, High Energy Density Phys. **24,** 44 (2017).

[18] J.-C. Pain, High Energy Density Phys. **26,** 23 (2018).

[19] M. K. Kruse and C. A. Iglesias, High Energy Density Phys. **31,** 38 (2019).

[20] H. R. Griem, A. C. Kolb, and K. Y. Shen, Phys. Rev. **116,** 4 (1959).

[21] M. Baranger, Phys. Rev. **112,** 855 (1958).

[22] J. W. Dufty, Phys. Rev. **187,** 305 (1969).

[23] R. Lee, J. Quant. Spectrosc. Radiat. Transfer **40,** 561 (1988).

[24] M. Baranger and B. Mozer, Phys. Rev. **123,** 25 (1961).

[25] W. Chappell, J. Cooper, and E. Smith, J. Quant. Spectrosc. Radiat. Transfer **10,** 1195 (1970).

[26] R. M. More, in *Laser Interactions with Atoms, Solids and Plasmas*, Vol. 327, edited by R. M. More (Springer US, Boston, MA, 1994), pp. 123–161.

[27] H. R. Griem, *Spectral Line Broadening by Plasmas*, Pure and Applied Physics Vol. 39 (Academic Press, New York, 1974).

[28] C. A. Iglesias, High Energy Density Phys. **6,** 318 (2010).

[29] C. A. Iglesias, High Energy Density Phys. **1,** 42 (2005).

[30] The perturbing electron interacts simultaneously with the bound electron and nucleus, so that the overall interaction consists of dipole and higher order terms.

[31] N. K. Rahman and F. H. M. Faisal, J. Phys. B **11,** 2003 (1978).

[32] M. S. Murillo and J. C. Weisheit, Phys. Rep. **302,** 1 (1998).

[33] S. Rose and R. More, in *Radiative Properties Of Hot Dense Matter, Proceedings of the 4th International Workshop, Sarasota, Florida*, edited by W. Goldstein, C. Hooper, J. Gauthier, J. Seely, and R. Lee (World Scientific Publishing Company, Singapore, 1991).

[34] A more general treatment may be retained in detailed calculations of individual line shapes, however, these are not tractable for the large number of lines present in opacity calculations.

[35] J. Cooper, Rev. Mod. Phys. **39,** 167 (1967).

[36] D. Kremp, M. Schlanges, W.-D. Kraeft, and T. Bornath, *Quantum Statistics of Nonideal Plasmas*, Springer Series on Atomic, Optical, and Plasma Physics No. 25 (Springer, Berlin, 2005).

[37] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.125.145002 which includes Refs. [36,38–40], for technical details of the electron-photon opacity calculation.

[38] M. F. Herbst, J. E. Avery, and A. Dreuw, Phys. Rev. A **99,** 012512 (2019).

[39] A. Dalgarno and J. T. Lewis, Proc. R. Soc. A **233,** 70 (1955).

[40] E. J. Robinson, Phys. Rev. A **58,** 755 (1998).