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A critical analysis of the advanced generalized theory: Applicability and applications

S. Alexiou[a], H.R. Griem[b,*], J. Halenka[c], W. Olchawa[c]

[a]*School of Physics, Department of Astronomy, Astrophysics and Mechanics, University of Athens, Athens, Greece*
[b]*Institute for Research in Electronics and Applied Physics, University of Maryland, College Park, MD 20742, USA*
[c]*Institute of Physics, Opole University, Olesca 4845-052, Opole, Poland*

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Abstract

A recent series of theoretical works (''advanced generalized theory'') has been proposed and applied to the analysis of hydrogen lines, particularly H_α in plasmas. The ''advanced generalized theory'' (AGT) [JQSRT 1994;51:129, Phys Rev E 1999;60:R2480, JQSRT 2000; 65:405] is critically examined, both theoretically and in applications to the analysis of experimental data. A number of serious flaws are exposed and discussed. The major flaws include using an inconsistent perturbation theory and erroneous Weisskopf radius-type arguments to access dynamic behavior. Further, the results derived from calculations using the theory are in disagreement with both exact analytic results and benchmark calculations giving rise to the conclusion that the theory is not physically valid. Finally, we find that applications of this theory to laser-spark and flash-tube experiments have led to claims of warm dense matter (WDM) effects, which are found here to be unnecessary when experimental errors are estimated realistically. In summary, we find the AGT to be incorrect in it formulation, incorrectly reduced to numerical results, and inappropriately applied to data analysis.

Keywords: Stark broadening; Hydrogen

*Corresponding author. Tel.: +1 301 405 4981; fax: +1 301 314 9437.
E-mail addresses: moka@hol.gr (S. Alexiou), griem@glue.umd.edu (H.R. Griem).

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1. Introduction

In recent years, the ''advanced generalized theory'' (AGT) [1–3] has been invoked to propose new high density effects for hydrogen line broadening calculations and claims have been made for agreement with various high density hydrogen experiments [1,2,4]. The AGT uses a parabolic basis and, as we shall illustrate, this and many other aspects of the formulation indicate that it is not general, in the sense of generally applicable. Indeed, more generalized approaches to spectral line broadening in plasmas exist in the literature and take many forms: the Pfennig–Lisitsa–Sholin (PLS) [5,6] approach which is an analytical, fully nonperturbative, solution of the time-dependent Schrödinger equation; the unified theory [7,8]; and the joint electron–ion simulations [9,10]. Further, we note that a critical measure of the validity of a theoretical formulation, which is found in all the theories mentioned as well as the standard theory (ST) [11,12], is a consistency in the assumptions and approximations employed. Here we will examine in detail the basic premises of, and results derived from, the AGT.

The AGT includes within its framework three components. The first of these components is the generalized theory (GT), which attempts to solve the impact broadening problem in the interaction picture with an unperturbed Hamiltonian consisting of the atomic Hamiltonian, the quasistatic ion microfield, using the Stark basis, and the component of the electron field along the quasistatic ion microfield direction. The second component is a residual ion impact width employed for all densities. The third component is a reduction of the electron impact broadening due to the acceleration of the perturber in the field of the nearest-neighbor ion to the emitter. We will analyze these components in the following sections.

2. Analysis of the original generalized theory (GT)

The idea behind the GT [1] is to solve the collisional broadening problem in the interaction picture with the unperturbed Hamiltonian consisting of the atomic Hamiltonian, the quasistatic ion field (i.e., the Stark basis), and the electronic field component in the direction of the quasistatic field. That is, the AGT essentially includes the ion field, taken to define the z-axis, plus the z-component of the electronic field in the unperturbed Hamiltonian. Perturbation theory is used for the x- and y- components of the electronic field. This leads to an inconsistency as all orders from the dressing factor associated with the z-component of the electric field are kept, but only the second order from the x and y-components. Furthermore, the convergence of this theory for the lateral Stark-split components of the line shape, though not for the central component, was taken as an indication that one could integrate down to impact parameter 0, neglecting a number of problems, such as unitarity and penetration. In particular, when penetration is taken into account, i.e., when the impact parameter becomes smaller than the atomic radial extent, it has been shown [13] that only slow collisions are strong, and fast collisions are not strong for any impact parameter. At zero impact parameter we have convergence for any velocity, while for fast enough collisions we have preservation of unitarity for all impact parameters. The reason for this convergence and unitarity preservation is penetration and not electron–ion coupling. Indeed, one obtains a convergent result even without any ions. Of course, convergence of a theory does not necessarily make it correct.

The main problem here is the transition from Eq. (9) to Eq. (11) in Ref. [1]. Since $E_z(t)$ involves the z-component of the electric microfield, we have in Eq. (9) a product of two expressions,

$$\exp\left(\mathrm{i}\Delta\, \mathrm{d}_z \int_{-\tau/2}^{\tau/2} \mathrm{d}t\, E_z(t)\right) \tag{1}$$

and

$$U_{\alpha\alpha}(\tau/2,-\tau/2)U^*_{\beta\beta}(\tau/2,-\tau/2), \tag{2}$$

where U is the upper/lower level U-matrix in an interaction representation for which the 0th order AGT Hamiltonian includes the atomic Hamiltonian, the quasistatic ion electric field as well as the time-dependent component of the electronic microfield along the direction of the quasistatic microfield. In Eq. (11) of Ref. [1] the second factor is expanded to second order:

$$U_{\alpha\alpha}(\tau/2,-\tau/2)U^*_{\beta\beta}(\tau/2,-\tau/2) \approx I - \int \mathrm{d}t_1 V'(t_1) \int \mathrm{d}t_2 V'(t_2). \tag{3}$$

The expansion of the above equation to second order in the interaction while keeping all orders of the potential in Eq. (1) is inconsistent and leads to unphysical contributions. The most problematic result of this inconsistent expansion is the appearance of a finite shift, whereas the shift is formally zero [14] as will be discussed below.

3. Analysis of the "residual ion contribution to the impact width (ICIW)"

The introduction of this component of the AGT first arises in Ref. [2], where it is suggested that for high densities and/or large principal quantum number (PQN) there is a residual ion impact width that increases with density and is independent of the PQN (at least for large PQNs). This suggestion led Oks [2] to question the validity of all simulations, as these allegedly assumed that the electron broadening dominates at high density and/or large PQN. The answer to this question is simply that the simulations, particularly joint electron–ion simulations [9], include all the physical processes in the AGT and no new effect has been found in the simulations. On the contrary, in the simulations it is clear that as the density and/or the PQN increase, the relevant time scale is reduced, so that in the very high density limit we have on average $\ll 1$ impact ion per configuration, since the number of relevant perturbers scales [9] as $T^{3/2}N^{-1/2}$ with T the temperature and N the density. Further, rare events are treated in simulations, but make no significant change in the line shape [9].

Let us emphasize that the ICIW is not a new physical effect, but simply an approximate way of computing a part of the ion dynamical contribution, which may currently be computed in its entirety with accuracy by a number of methods [15,16]. We find that there are three basic assertions underlying the ICIW that are incorrect:

First, there is the assertion that this ICIW at high densities and/or PQN can only be obtained within the GT and, specifically, that it is not obtainable with the ST. On the contrary, we will

illustrate in the Appendix A that the ST produces an ICIW, whereas the GT formally produces *no such width*. This latter fact is due to the adiabatic contribution arising from a *one-dimensional* field, which by itself is unable to broaden the central component, as in the case of quasistatic broadening. This is further discussed in Appendix B.

Second, the ICIW proposed in Ref. [2] is claimed to be applicable to all transitions; however, this can be shown to be incorrect. Indeed, the entire discussion of the ICIW is relevant only for lines with a central component, for which quasistatic broadening is ineffective. The proof of this is shown in Appendix A.

Third, there is the assertion that the ICIW can be comparable to the standard electron impact width (EIW). Note that the ICIW is simply an approximate way to estimate the ion impact width, which in turn is *a part* of the ion dynamical contribution, which itself is *a part* of the ionic contribution. In Appendix A we show that an analytical calculation of the ICIW seriously overestimates the ion impact contribution. We also explain why this overestimation is built into the analytical calculation of the ICIW. The analytical calculation of the ICIW, on the one hand, overestimates the ion impact contribution and, on the other hand, completely neglects the dynamic but nonimpact ionic contribution (as does the ST). The point is that the ICIW is not a new effect, but simply a part of that part of the ionic contribution which has long been known to exist for any plasma parameters [17]. It has also been well understood for lines with a central component that in the high density limit, EIW dominates all ion dynamical contributions by virtue of its linear density scaling. It has also been well understood that at low enough densities ion impact dominates EIW, as well as that ion dynamics, of which ion impact may be an important part, remains important over a substantial range [18,17]. Specifically, the EIW [12] to ICIW ratio may be estimated as

$$\frac{EIW}{ICIW} \approx 0.0323\left(\frac{N_e}{10^{18}\mathrm{e/cc}}\right)^{2/3} Z_i^{1/3} T_{ev}^{-1} \ln\left(\frac{\lambda_D}{\rho_{min}}\right)(n_u^2 - n_l^2)^2. \tag{4}$$

Here T_{ev} is now the temperature (assuming equal electron and ion temperatures) in eV, Z_i the ionic charge and N_e the electron density. Also, the Debye radius λ_D and ρ_{min} are the maximum and minimum impact parameters. This can be less than or equal to unity for low-lying lines (L_α, H_α), high T (but note $T \leqslant 10\,\mathrm{eV}$ for hydrogen) and even for the low density, high n case discussed in [2]. Qualitatively these are precisely the conditions for ion dynamics to be important. On the other hand, in the very high density and/or high PQN limit the ICIW is negligible compared to the EIW, as expected. In that limit the importance of the nonimpact ion part also grows relative to the ICIW, since shorter time scales are involved over which the ion field is smoother. In short, the occurrence of an ion impact width contribution at high densities is neither new, nor unexpected. It is well known that the criteria for the applicability of the quasistatic approximation are not clear-cut for lines with a central component and high temperatures [18], so that researchers have learned not to assume static ions in such cases.

We do *not* propose using the ICIW as computed in Appendix A, because there are more general and accurate ways currently available of dealing with *all* of ion dynamics. Also, in Appendix A we test our analytical ICIW against exact calculations which are free from the assumptions in the ICIW derivation. These exact calculations are used in Appendix A to test these assumptions. As

already mentioned, we find that the ICIW overestimates the ion impact contribution, for example, by 50% in one case studied in Appendix A.

In summary, the problems with the proposed concept of an ICIW are:

First, there is no "stand-alone" adiabatic width contribution for lines with a central component. That is, the ICIW is identically zero in the AGT.

Second, a correct formulation of the ICIW can be obtained within the ion impact ST, so it cannot be considered a "new" effect.

Third, the analytic ICIW is of limited accuracy, but still in many cases much better than the static ion assumption.

Fourth, in the high density and/or PQN limit, electron broadening dominates the ICIW, as understood from all previous formulations.

4. Analysis of the acceleration by the nearest neighbor

The last component of the AGT is a narrowing effect attributed [3] to the acceleration of the perturbing electron in the field of the nearest-neighbor ion to the emitter (AEIF). For a neutral emitter this means that the perturbing electron straight path is transformed to a hyperbolic path. This is, of course, in contrast to the normal picture of noninteracting quasiparticles (for instance, see Ref. [9]), but the idea is of interest in principle because at high densities the interactions between these particles could conceivably have an effect. Within the Debye sphere it is known that one can treat the perturber dynamics in a detailed manner. However, although questions regarding the limits of temperature, density and ion charge where the perturber–perturber interaction becomes important in spectral line broadening are interesting, it can be verified that static properties effecting the line profile are not appreciably modified at the plasma parameters at which this effect has been proposed [3] to be important. That is, for static properties the assumption that perturbers are noninteracting quasiparticles is practically as accurate as including their correlations via the APEX [19] microfield distribution.

Moreover, the AEIF effect may be overestimated in Ref. [3], for two reasons: (1) The incorrect assumption or misprint in Eq. (3) of [3], that the electron impact broadening scales essentially as $1/v^2$ rather than $1/v$; and (2) the use of an incorrect energy balance for the perturbing electrons that ignores Debye shielding, see Eq. (1) in Ref. [3]. In fact, ST calculations and computer simulations show [20] that a more realistic estimate of any AEIF effect differs from the result of Ref. [3] both in magnitude and direction, i.e., we obtain an increase of 3–15% in lieu of the ~25% decrease in the widths of H-alpha predicted by Oks.

5. Analysis of the applications of the AGT to experimental data

First, note that the GT is at odds with well-established theoretical models [21,22]. For example, it is claimed that in the Grützmacher–Wende [23] experiments ion dynamics has been overestimated [24]. These experiments have been found to agree with benchmark simulations [9], which include all the electron–ion coupling included in the GT and, most importantly, include additional contributions that are beyond the impact-quasistatic approximations, binary approximation and independent electron–ion

broadening approximation. These benchmark calculations treat the full joint electron–ion microfield exactly and, furthermore, find no electron–ion coupling; that is, the computed line profile is in excellent agreement with the profile obtained by convolving the electronic and ionic profiles.

Second, there is the claim, (see Ref. [25]), that the AGT is verified experimentally. However, the ST calculations are also within the experimental error bounds [20,26]. Thus, we find that in the cases where the AGT has been applied to data analysis there are substantial inconsistencies with both existing validated calculational approaches and the attribution to new effects that under correct analysis are within the experimental errors. In addition, the AGT also needs a strong collision term, which may be determined in the high density, low temperature regime only with far larger error bounds than ST–AGT differences.

It has been shown in Refs. [27,28] that serious errors were made in application and analysis of data presented in Escarguel et al. [29]. In particular, the electron densities N_e and temperatures obtained in [29] are unreliable, because for such physical conditions the investigated plasmas would have densities about 1.5 times higher than *normal water*, which is not possible. Thus, the FWHM and/or shift versus N_e relations reported in [29] must be considered incorrect; however, inspite of this fact agreement between the AGT and the experiment is cited, which can be considered as further proof that the AGT yields incorrect results.

6. Summary and conclusions

We find that the proposed AGT is incorrect both in its formulation and in the reduction to calculations. To summarize the problems:

First, because the AGT uses an inconsistent perturbation theory, it predicts nonzero dipole shifts for hydrogen lines in the no-quenching approximation and with the usual density matrix factorization [30]. This has been proven incorrect both numerically [27] and analytically [14].

Second, the AGT incorporates a residual impact ion width ICIW that we have shown is incorrectly calculated.

Third, the AGT incorporates the acceleration by the nearest neighbor, AEIF, but the effect is incorrectly estimated for numerous reasons, for example [20], using an overly simplified model, neglect of correlations and the neglect of the electron perturbation by the nearest neighbor.

Fourth, the AGT has been applied to the analysis of various experiments and in each a new effect has be proffered to explain the data. We have shown that these claims are unfounded both because the AGT is at its foundation invalid and because an experimental error analysis indicates that new effects are not needed to explain the data.

In conclusion, we firmly believe that the AGT is seriously flawed and great care must be exercised when results derived from it are used.

Appendix A. ST ICIW computation and analysis

In the GT the (adiabatic) impact width is given by the expression

$$HWHM = 2\pi N \int \mathrm{d}v\, v f(v) \rho_w^2(v) \int_0^{L/\rho_w} z\, \mathrm{d}z \left(1 - z \sin\frac{1}{z}\right) \tag{5}$$

with N the density of the species of interest, i.e., here the impact ions, and L a maximum impact parameter that is the minimum of the screening length and a length for which the contribution can be considered as impact, as we discuss below. Note that the z-integrand is $\sim z$ for $z \leqslant 0.3$ and $\sim(6z)^{-1}$ for $z \geqslant 1$, which may be interpreted as the strong vs. weak collision regimes. In Ref. [2] it is implicitly assumed that the relevant z are $z \ll 1$, which may be justified a posteriori, so that the small z-limit of the $1 - z \sin 1/z$ expression may be taken.

At this point we note that the ST also predicts an ICIW, except it does so without associating a width with a one-dimensional field. Furthermore, whereas in Ref. [2] the ICIW is a large PQN and/or density limit result, here this is quantified more precisely. If R is a length such that all collisions with smaller impact parameters are *both* strong and impact, then we get for the ST

$$HWHM = 2\pi N \int \mathrm{d}v\, v f(v) \int_0^R \{1 - S_a S_b^{\dagger}\} \rho \,\mathrm{d}\rho \approx 2\pi N \int \mathrm{d}v\, v f(v) \int_0^R \rho \,\mathrm{d}\rho, \tag{6}$$

because for strong collisions $\{1 - S_a S_b^{\dagger}\}$ oscillates around unity.

To make sure this width is due to impact ions alone, we need to choose a characteristic frequency Ω and include only those perturbers with $v/\rho > \Omega$ in such a way that $HWHM(\Omega) = \Omega$, where $HWHM(\Omega)$ denotes the width due to only these ions [17].

Then we have replacing R^2 by $(v/\Omega)^2$ for $v \lesssim R\Omega$:

$$HWHM(\Omega) = \pi N \left[\Omega^{-2} \int_0^{R\Omega} \mathrm{d}v\, v^3 f(v) + R^2 \int_{R\Omega}^{\infty} \mathrm{d}v\, v f(v) \right]. \tag{7}$$

With μ denoting the reduced ion mass, this is simply evaluated to [17]

$$HWHM(\Omega) = \pi N R^2 \sqrt{\frac{8kT}{\pi\mu}} F\left(\frac{\mu(\Omega R)^2}{2kT} \right) \tag{8}$$

with

$$F(x) = \frac{2(1 - \mathrm{e}^{-x}(1 + x + x/2))}{x} + (1 + x)\mathrm{e}^{-x}. \tag{9}$$

$F(x)$ is plotted in Fig. 1. Note that $F(x) = 1$ corresponds to an impact contribution from the entire phase space ($\Omega = 0$).

In the above HWHM expression, the only Ω-dependence is found in F. If the argument of F is small, so that $F[\mu(\Omega R)^2/2kT] \approx 1$, the width is Ω-independent. The condition for the impact approximation to be valid is then

$$HWHM(\Omega) = \Omega = \pi N R^2 \sqrt{\frac{8kT}{\pi\mu}}. \tag{10}$$

If we accept that $[\mu(\Omega R)^2/2kT] = a$, $F(a) \approx 1$ (hence, a is the point at which we say that $F(x)$ is still approximately 1, i.e., $a \lesssim 1$), we have

$$\Omega = R^{-1} \sqrt{\frac{2akT}{\mu}} = \pi N R^2 \sqrt{\frac{8kT}{\pi\mu}}, \tag{11}$$

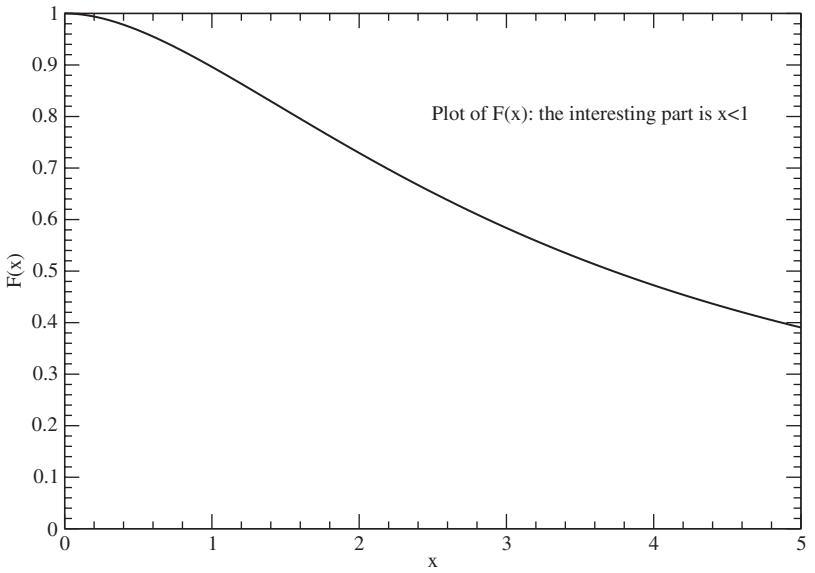

Fig. 1. Plot of $F(x)$.

which allows us to solve for R:

$$2\pi NR^3 = \sqrt{\pi a}. \tag{12}$$

This means that R is indeed proportional to the mean interionic spacing with a proportionality constant of the order of 1. This means that $\{1 - S_a S_b^{\dagger}\} \approx 1$ was a reasonable assumption if R is small enough to invalidate a perturbative expansion (i.e. smaller than the so-called Weisskopf radius ρ_w).

Numerically, some remaining issues are the choice of a (which in the GT case corresponds to the question "what is the maximum z for which $z[1 - z\sin(1/z)] \approx z$"), and the temporal variation of the field of a perturber with impact parameter ρ and velocity v is taken in Ref. [2] to be $3^{-1/2}\rho/v$. In that case, the HWHM needs to be equal to the inverse of the field temporal variation, i.e., $\sqrt{3}\Omega$, and R is determined similarly.

So we have obtained the ICIW using standard ST arguments and two assumptions: strong collisions ($\{1 - S_a S_b^{\dagger}\} = 1$) and the impact criterion being $HWHM(\Omega) = \Omega$. We now compare the ICIW with an exact calculation for the H_α line. For the exact calculation we perform a simulation for a temperature of 8.4 eV and an electron density of 4.84×10^{18} e/cm^3 and helium ions. In these simulations we artificially zeroed the fields of all ion perturbers with impact parameters ρ and velocities v satisfying $v < \Omega\rho$ for different choices of Ω. We denote the resulting width HWHM(Ω). In this way we obtained the "impact" boundary Ω from the equation $HWHM(\Omega) = \Omega$ and illustrate this graphically in Fig. 2.

This procedure results in $\Omega = 12.4 \times 10^{12}\,\mathrm{s}^{-1}$ and is about 50% lower than the ICIW estimate from [2], which was $18 \times 10^{12}\,\mathrm{s}^{-1}$. This HWHM width of $12.4 \times 10^{12}\,\mathrm{s}^{-1}$ from impact ions, i.e. about 30 Å, is to be compared with a total (electrons and ions) ST FWHM width of about 160 Å, with electron broadening amounting to about 60 Å. Although simply adding the impact width to a

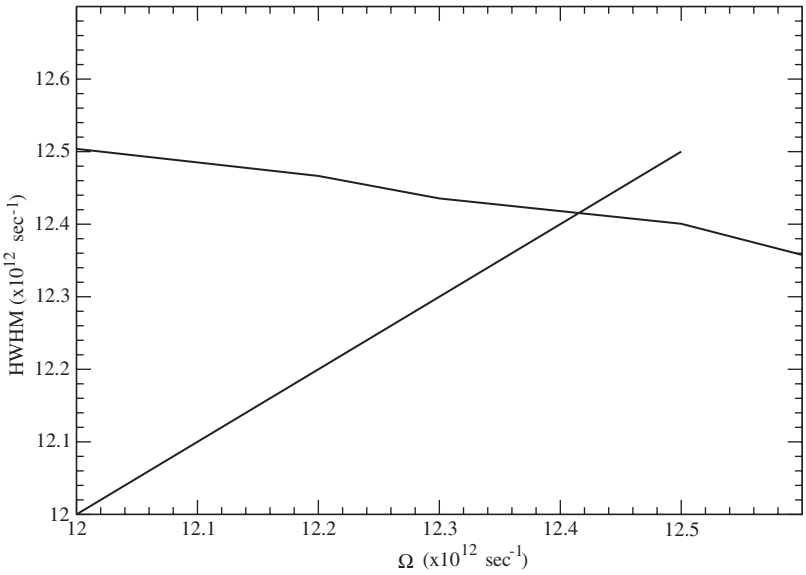

Fig. 2. Graphical determination of the angular frequency Ω separating impact from nonimpact ions for H_α.

nonimpact width is theoretically not justified, since the impact and nonimpact components are comparable here, in practice [16] such coupling effects are small.

The above comparison shows that the ICIW is larger than exact computation of the effect and we now investigate the reasons for these discrepancies.

To check the two assumptions in the analytic computation of the ICIW we show in Fig. 3 the quantity $Q(t) = -[9/4][eE_k(t)a_0/\hbar]$ $(k = x, y, z)$ in ps^{-1} vs. time for two different configurations. Both configurations were generated subject to the requirements that $v/\rho \geqslant HWHM$ and the time of closest approach is less than a few times the inverse HWHM (and $\geqslant 0$). The first (solid lines) configuration is a typical impact configuration, with a small strong collision duration on the inverse HWHM time scale. The second (dashed lines) not only has a duration that is more comparable to the inverse HWHM time scale, but also exceeds the inverse HWHM time scale. Although collisions with impact parameters up to distances $\rho_w > R$ are strong, for small velocity collisions their fields may be smoothed out, whereas very fast collisions cannot be smoothed out and appear as sharp, impact components.

If we solve the equation $\Omega = 2HWHM(\Omega)$, meaning that one only considers impact perturbers with a time variation at least twice as fast as the inverse HWHM time scale, we find instead an impact width of $9 \times 10^{12}\,s^{-1}$, compared to $12.3 \times 10^{12}\,s^{-1}$ for $\Omega = HWHM(\Omega)$. This means that such perturbers, which are marginally impact, are important to the ICIW calculation. Also, collisions, even strong collisions, with collision times $\geqslant HWHM^{-1}$ are not as effective, because other broadening mechanisms (nonimpact ions, electron impact) have already produced a significant decay of $C(t)$, as demonstrated in Fig. 4.

Last, in Ref. [2] the ICIW is presumably used for any line, e.g., $D_8 - D_{11}$. However, a separate additive broadening mechanism consisting of strong impact collisions is invalid for lines without a central component. Such lines typically exhibit a central local minimum or dip. In terms of $C(t)$

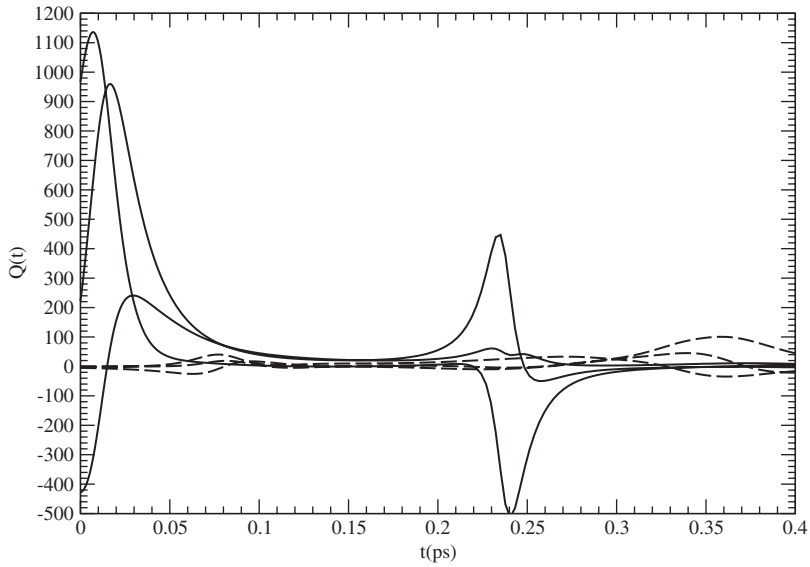

Fig. 3. Fields for two configurations.

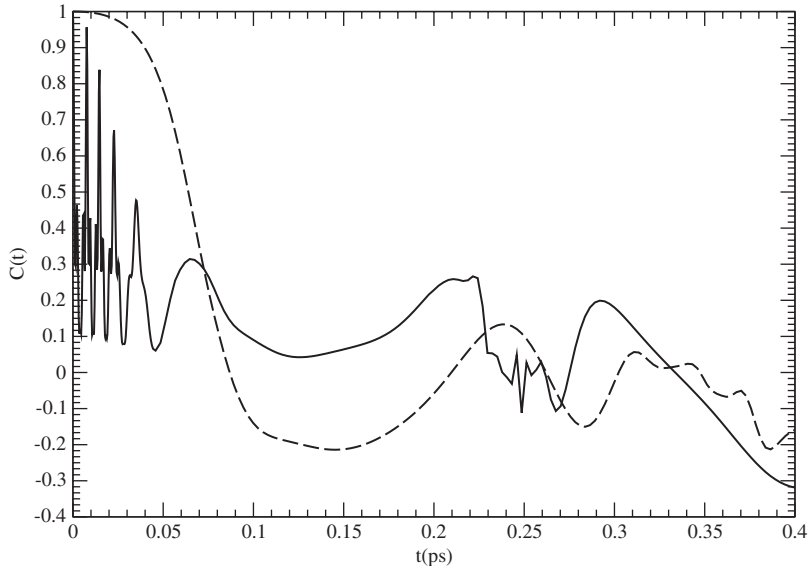

Fig. 4. Contributions to the autocorrelation functions for the two configurations.

this dip translates into a region where $C(t)$ goes negative. The effect of (ion or electron) dynamics is to make this negative $C(t)$ narrower and hence to fill in the dip. Adding widths is appropriate for Lorentzian profiles, but here the profile is not Lorentzian and adding an impact width to the quasistatic width is conceptually incorrect. This is another subtlety that cannot be derived using a Weisskopf radius argument.

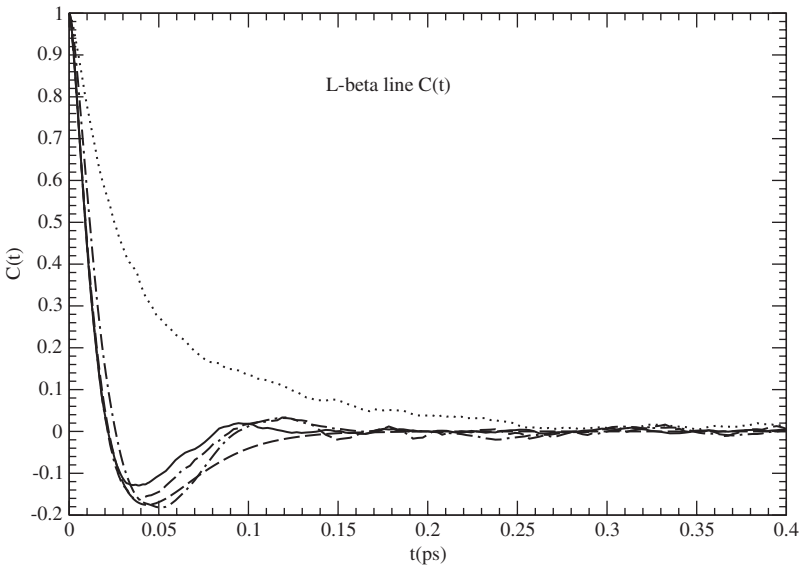

Fig. 5. $C(t)$ Illustration for a line without a central component.

To illustrate this point, let us consider the L_β line at the same parameters as before, i.e., at a temperature of 8.4 eV and electron density of 4.84×10^{18} e/cm^3, with He^+ perturbers. In that case (and neglecting electrons, as before), the equation $HWHM(\Omega) = \Omega$ gives $\Omega \approx 17 \times 10^{12}\,s^{-1}$. This is about one-seventh of the total width $11.16 \times 10^{13}\,s^{-1}$.

In Fig. 5 we show the autocorrelation functions for (a) all ions (solid line), (b) all ions in the quasistatic approximation (dashed line), (c) only impact ions (dotted line), (d) only nonimpact ions (dash-dotted line) and (e) nonimpact ions treated dynamically and impact ions treated as if they were quasistatic (i.e., with their field frozen at the value they had at $t = 0$) (double dash-dotted line). Note that the differences between a, b and e are only present for the negative region, i.e., the region responsible for the dip. In Fig. 6 we replot the dynamic (solid) and quasistatic (dashed) autocorrelation functions, and we also show their difference (dotted). Note that this difference is a *localized* peak with a center at about 0.07 ps.

In other words, it makes very little difference at all for the width whether one treats these impact ions as quasistatic, or whether one computes them rigorously. The fact that (ion or electron) dynamics do not contribute much to the width, but mostly to the filling in of the dip, is well known for many years in the literature [31]. We note that for D_8 in the example used in [2], the ICIW amounts to 0.7 Å, i.e., about 15% of the measured FWHM. The application of the ICIW to lines without a central component is yet another error in the AGT.

Appendix B. Proof that the GT ICIW is strictly zero

Below, we first explain why the ICIW in GT is *exactly* zero: the proposal in Ref. [2] uses an inappropriate argument to associate strength of collision, due to impacts for $D/\rho_w \gg 1$ and

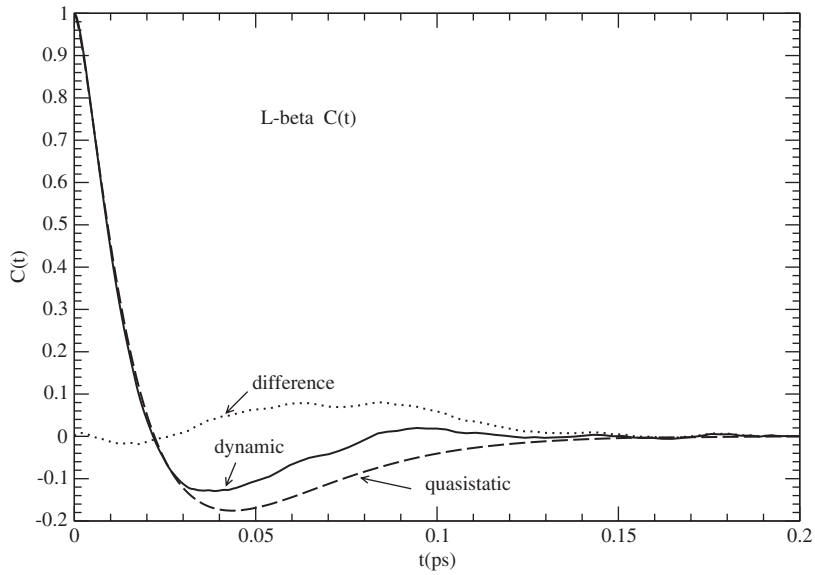

Fig. 6. Difference of dynamic and quasistatic $C(t)$ for L_β at a temperature of 8.4 eV and electron density of 4.84×10^{18} e/cc, with Helium perturbers.

dynamical behavior, due to quasistatic effects for $D/\rho_w \ll 1$ with D the mean interionic distance and ρ_w the Weisskopf radius. These Weisskopf radius arguments are understood [31,21] to be incorrect when assessing static/dynamic behavior, as they refer to the strength of the interaction, not the dynamic effects. As an example we illustrate that the Weisskopf radius criterion is not a reliable one for accessing dynamic behavior, because its application to electrons leads to the unphysical result that at high densities electrons are also static—a nonsensical proposal. Moreover, we note that for the case of lines with central components the "dominant" adiabatic contribution for these impact ions is computed in Ref. [2] using the z-component alone, i.e., only the field component that is along the quasistatic field. However, *any one-dimensional field cannot broaden the central component*. This is well known and easily seen in either spherical or parabolic coordinates: in spherical coordinates, the perturbation of the maximum $|m|$-states due to z-matrix elements is 0, e.g.,

$$\mathrm{d}\langle n,l,m|U(t)|n,n-1,n-1\rangle/\,\mathrm{d}t \sim -\frac{\mathrm{i}}{\hbar}\langle n,l,m|z|n,l',m'\rangle\langle n,l',m'|U(t)|n,n-1,n-1\rangle = 0. \tag{13}$$

At $t=0$, $l'=n-1=m'$, and the z-matrix element is identically 0, so that there is no U-matrix element involving states that give rise to the central component. The point is that an adiabatic contribution alone will lead to *δ-function* central components. This is similar to the quasistatic contribution as it can only enhance the impact broadening of a central component and not produce a width by itself. Therefore, Eq. (1) of Ref. [2] is incorrect: the residual ion impact width, as computed there, is zero for lines with an unshifted component.

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