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AN ANALYSIS OF MARKOVIAN MODEL MICROFIELD METHODS FOR STARK BROADENING

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Abstract—Stark broadening theories which concentrate on the statistics of the plasma electric microfield rather than the dynamics of collisions have come to be known as Model Microfield Methods. In the present paper we present an analysis of Stark broadening problems based on Markovian model microfield statistics. Our derivation permits an easy comparison with traditional Stark broadening theories such as the impact and unified theories; this comparison is used to clarify the physical nature of the approximations employed by Markovian models. The strengths and weaknesses of various models are discussed, emphasizing the kangaroo process of Brissaud and Frisch, and methods are suggested for improving the current model microfield approach.

1. INTRODUCTION

In this paper we will discuss the class of line broadening theories known as Model Microfield Methods[1–4] henceforth abbreviated as MMM. This type of theory was originally proposed by Brissaud and Frisch[1] for line broadening in plasmas where a radiating atom is perturbed by the electric microfield produced by the electrons and ions. In traditional theories, such as the impact[5] and unified[6,7] theories, the electric microfield is evaluated as the sum of electric fields produced by each of the charged particles in the plasma and averages of various collisional operators such as time development operators are performed by independently averaging over all possible positions and velocities for the charged particles. The MMM proposed by Brissaud and Frisch makes a radical departure from the traditional approach because it concentrates on the statistical properties of the electric microfield, almost disregarding the existence of the particles. The goal of the MMM is to find a mathematical model which correctly describes those statistical properties of the electric microfield which are necessary for calculating line profiles. A few key parameters are of course defined by results obtained from the traditional plasma kinetic theories, but the main focus of the theory is on the mathematical statistics.

The approximations used by Brissaud and Frisch are consequently mathematical in nature rather than physical and they were motivated in part by the desire to obtain a simple analytic expression for the line shape. It has thus been quite difficult to assess the validity of their model and the MMM approach was, to a large extent, ignored for this reason. However, Seidel[3] has recently performed a detailed set of calculations for hydrogen lines using the MMM of Brissaud and Frisch to treat the electrons and these calculations agreed very closely with the best available unified theory calculations. Shortly thereafter, he used the MMM for the ions as well[4] and these calculations gave much better agreement with experimental data then all previous impact and unified theory calculations. Nevertheless, in spite of this improvement, there are still discrepancies with experimental data. As a result of the success of Seidel's calculations and the tantalizing discrepancies which remain, there is now a great deal of interest in the MMM, its conditions for validity, and methods for improving the model.

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The goals of the present paper are to (1) present a new approach to the MMM which permits a detailed comparison with the traditional impact and unified theories, (2) to clarify the physical nature of the approximations used by Brissaud and Frisch, (3) to point out weak points in the theory which may limit its validity and (4) to suggest methods for improving MMM calculations.

2. DIGRESSION ON STRONG COLLISIONS

We wish to begin with a brief digression on the difference between strong dynamic collisions and strong static interactions. This is necessary because we will be interested in discussing the validity of various theories for strong fields and also because a great deal of confusion exists in the literature concerning the distinction between strong dynamic and static fields.

The traditional view of static interactions begins by noting that the intensity at a frequency separation $\Delta\omega$ from line center is given by the Fourier transform of the dipole autocorrelation function

$$I(\Delta\omega) \propto Re \int_0^\infty e^{i\Delta\omega t} \langle \mathbf{d}(t) \cdot \mathbf{d}(0) \rangle. \tag{1}$$

This integral is determined primarily by the region $t \leq 1/\Delta\omega$ because, for $t > 1/\Delta\omega$, the exponential oscillates between -1 and 1, which reduces the magnitude of the integrand and produces a cancellation between positive and negative values. Letting τ denote the duration of a collision between a charged particle and the radiating atom, it is then clear that all particles for which $\tau \gg 1/\Delta\omega$ may be regarded as stationary when calculating the intensity at the point $\Delta\omega$. Thus, it is argued that for $\Delta\omega$ sufficiently large, most particles could be regarded as static. Letting $V(r)$ denote the first order static perturbation[8] due to a particle at the point r, the energy levels of the atom will be shifted so that it will absorb or emit at a new frequency ω defined by $\hbar\Delta\omega = V(r)$. Combining this result with the requirement $\tau \gg 1/\Delta\omega$ for static interactions, one finds that static interactions are characterized by

$$V\tau/\hbar \gg 1 \text{ and } \Delta\omega \gg 1/\tau \quad \text{(static interaction)}; \tag{2a}$$

if $V(r) = C/r^n$ with $n \geq 2$ and $\tau = r/v_{\text{avg}}$, where v_{avg} is the average perturber velocity (relative to the atom), then Eq. (2a) gives

$$r \ll r_w \equiv (C/\hbar v_{\text{avg}})^{1/(n-1)} \text{ and } \Delta\omega \gg 1/\tau \quad \text{(static interactions)}; \tag{2b}$$

where r_w is called the Weisskopf radius; a corresponding Weisskopf frequency is defined by the inverse of the collision duration $\tau_w = r_w/r_{\text{avg}}$, that is $\Delta\omega_w = v_{\text{avg}}/r_w$. Thus, one says that static interactions are characterized by Eq. (2a) and, on the average (i.e. using $v = v_{\text{avg}}$), they are due to perturbers which are much closer than the Weisskopf radius r_w and they contribute to that region of the linewings where $\Delta\omega \gg \Delta\omega_w$ (i.e. using $\tau \ll \tau_w \equiv 1/\Delta\omega_w$ and the condition for static interactions $\tau \gg 1/\Delta\omega$).

On the other hand, strong collisions are defined to be all those interactions for which a perturbation expansion of the S matrix is not valid. That is,

$$\hbar^{-1} \int_{-\infty}^{\infty} V(t)\, dt \cong \tau V(r)/\hbar \geq 1 \quad \text{(strong interactions)}, \tag{3}$$

where r is the distance of closest approach and $\tau \cong r/v$ is the collision duration time; using the Weisskopf radius defined in Eq. (2b), this is equivalent to

$$r \leq r_w \quad \text{(strong interactions)}. \tag{4}$$

Comparing Eqs. (2a) and (3) or (2b) and (4), we see that all static interactions are strong *but the converse is not true.* In the line center, $\Delta\omega \lesssim 1/\tau$, all strong collisions will be dynamic and, even in the line wings, $\Delta\omega \gg 1/\tau$, the interactions for which $1 \lesssim V\tau/\hbar \lesssim 10$ may be regarded as dynamic strong collisions. It is important to realize that one may not treat all strong interactions as though they are static; a failure to appreciate this fact (see Section 2.1 of Ref. 9) has led

Brissaud *et al.*[9] to an incorrect assessment of the validity of their MMM for strong interactions (this will be discussed further in Section 5).

In order to give a rough but quantitative description of these concepts, we consider low lying hydrogen lines (e.g. Ly-α, Ly-β, H_α, H_β) in a plasma at a temperature $T = 10^4$ K and density $n = 10^{17}\,\mathrm{cm}^{-3}$ as an example. A reasonable estimate for $V(r)$ is $V(r) \cong 10a_0e^2/r^2 \cong 1.2 \times 10^{-26}\,\mathrm{erg\,cm^2}/r^2$ for $\lambda_{th} \leq r \leq \lambda_D$. For r greater than the Debye length $\lambda_D = \sqrt{(kT/4\pi ne^2)}$, the interaction is cut off by plasma shielding effects and, for r smaller than the thermal de Broglie wavelength $\lambda_{th} = \sqrt{(h/2\pi\mu kT)}$, classical concepts are no longer valid ($r < \lambda_{th}$ contributes at most $\pi\lambda_{th}^2$ to the optical cross section which is a small correction for most plasmas). Noting that the reduced mass μ for a hydrogen–proton collision is roughly half the proton mass, the various parameters of interest are

$$\begin{aligned}\lambda_{th} &= \hbar/\sqrt{(2\pi\mu kt)} \\ &= 7.45 \times 10^{-8}\,\mathrm{cm}\,\sqrt{(10^4/T(^\circ K))} \quad \text{(electrons)} \\ &= 2.46 \times 10^{-9}\,\mathrm{cm}\,\sqrt{(10^4/T(^\circ K))} \quad \text{(ions)},\end{aligned} \tag{5}$$

$$\begin{aligned}\lambda_D &= \sqrt{(kT/4\pi ne^2)} \\ &= 6.9 \times 10^{-7}\,\mathrm{cm}\,\sqrt{(T(^\circ K)/10^4)}\,\sqrt{(10^{18}/n(\mathrm{cm}^{-3}))},\end{aligned} \tag{6}$$

$$\begin{aligned}r_w &= (C/\hbar)\sqrt{(\mu/3kT)} \\ &= 1.7 \times 10^{-7}\,\mathrm{cm}\,\sqrt{(10^4/T(^\circ K))} \quad \text{(electrons)}, \\ &= 5.16 \times 10^{-6}\,\mathrm{cm}\,\sqrt{(10^4/T(^\circ K))} \quad \text{(ions)}.\end{aligned} \tag{7}$$

Thus we see that, for ions at $T = 10^4$, $N = 10^{17}$ $r_w \gg \lambda_D$; hence, all interactions are strong; some will be static and some dynamic depending on the value of $\Delta\omega$. For electrons, $r_w \cong 2\lambda_{th}$; hence, there will be very few strong dynamic collisions and essentially no static collisions (i.e. they all fall in the quantum scattering domain $r \lesssim \lambda_{th}$ which makes very little contribution in most cases).

We next note that the probability finding two perturbations simultaneously within a distance r from the radiator is $(4\pi r^3 n/3)^2 = (r/r_0)^6$ where $r_0 \equiv (3/4\pi n)^{1/3} = 6.2 \times 10^{-7}\,\mathrm{cm}\,(10^{18}/n)^{1/3}$. Taking $r \lesssim r_w$, this procedure gives the probability of having two simultaneous strong collisions. For electrons, $r_w \ll r_0$ and one may neglect the problems of overlapping strong collisions; for ions on the other hand, $r_w \cong 10r_0$; hence, the overlap of strong interactions will be a very important part of ion broadening.

3. DERIVATION OF MARKOVIAN MODELS FOR THE ELECTRIC FIELD

In this section, we outline the mathematics which provides the foundation for any Markovian model of the plasma electric field, following closely the discussion of Markov processes contained in Ref. 10. We also derive various expressions for the line shape which permit an analysis of the validity of the MMM. The kangaroo process (KP) proposed by Brissaud and Frisch[1,2] is discussed as a specific type of Markovian model.

We start by considering a series of values $(\epsilon_1, \epsilon_2, \epsilon_3, \ldots, \epsilon_n)$ taken by the electric field $\epsilon(t)$ at the times $(t_1, t_2, t_3, \ldots, t_n)$. The probability of finding these values at the prescribed times is described by a probability function $\Phi_n(\epsilon_1 t_1; \epsilon_2 t_2; \epsilon_3 t_3; \ldots; \epsilon_n t_n)$. These changes in $\epsilon(t)$ are said to be a Markov process whenever[10]

$$\begin{aligned}\Phi_n(\epsilon_n t_n; \epsilon_{n-1} t_{n-1}, \ldots, \epsilon_2 t_2; \epsilon_1 t_1) &= P(\epsilon_n t_n | \epsilon_{n-1} t_{n-1}) \\ &\quad \times P(\epsilon_{n-1} t_{n-1} | \epsilon_{n-2} t_{n-2}) \ldots P(\epsilon_2 t_2 | \epsilon_1 t_1) \Phi(\epsilon_1 t_1),\end{aligned} \tag{8}$$

where $P(\epsilon_2 t_2 | \epsilon_1 t_1)$ is a conditional probability, that is, it gives the probability that the electric field $\epsilon(t)$ will have the value ϵ_2 at the time t_2 when it is known that $\epsilon(t_1) = \epsilon_1$.

For a Markov process, the conditional probability function satisfies the Chapman–Kol-

mogoroff equation[10]

$$P(\epsilon t|\epsilon' t') = \int d\epsilon'' P(\epsilon t|\epsilon'' t'') P(\epsilon'' t''|\epsilon' t') \tag{9}$$

as well as the following relations:

$$\int d\epsilon' P(\epsilon t|\epsilon' t') = 1, \tag{10}$$

$$\int d\epsilon' P(\epsilon t|\epsilon' t') \Phi_1(\epsilon' t') = \Phi_1(\epsilon t). \tag{11}$$

We next restrict ourselves to a subclass of Markov processes for which[11] [see Eq. (I.73) of Ref. 10 or Eq. (3.12) of Ref. 2]

$$P(\epsilon t + \Delta t|\epsilon' t) \xrightarrow[\Delta t \to 0]{} A\delta(\epsilon - \epsilon') + \Delta t W(\epsilon|\epsilon'), \tag{12}$$

where $W(\epsilon|\epsilon')$ is called the transition rate, A is a normalization constant required by Eq. (10), viz.

$$A = 1 - \Delta t \int d\epsilon' W(\epsilon'|\epsilon) = 1 - \Delta t \nu(\epsilon), \tag{13}$$

and $\nu(\epsilon)$ is a frequency defined by

$$\nu(\epsilon) = \int d\epsilon' W(\epsilon|\epsilon) \tag{14}$$

such that $1/\nu(\epsilon)$ is the average duration of a field of strength ϵ. We note in passing that Eq. (12) implies that

$$P(\epsilon t|\epsilon' t) = \delta(\epsilon - \epsilon'). \tag{15}$$

Substituting Eq. (12) into Eq. (9), we obtain the differential form of the Chapman–Kolmogorov equation[10]

$$\partial_t P(\epsilon t|\epsilon' t') = -\nu(\epsilon) P(\epsilon t|\epsilon t') + \int d\epsilon'' W(\epsilon|\epsilon'') P(\epsilon'' t|\epsilon' t'), \tag{16}$$

which is better known in physics as a master equation. In fact, multiplying Eq. (16) by $\Phi_1(\epsilon' t)$, integrating over ϵ' and using Eq. (11) gives the familiar master equation for Φ_1

$$\partial_t \Phi_1(\epsilon t) = -\nu(\epsilon) \Phi_1(\epsilon t) + \int d\epsilon'' W(\epsilon|\epsilon'') \Phi_1(\epsilon'' t). \tag{17}$$

Starting from Eq. (16), Brissaud and Frisch define a specific Markov process, which they have named the kangaroo process, by choosing a transition rate W with the product form

$$W(\epsilon|\epsilon') = Q(\epsilon)\nu(\epsilon'). \tag{18}$$

There are many ways to specify the functional form of $Q(\epsilon)$ and $\nu(\epsilon)$ and the functions chosen by Brissaud and Frisch are well adapted to line broadening problems. They choose the function Q so that the steady state solution of the master equation, Eq. (17), will be the well known plasma microfield distribution function[12] $P(\epsilon)$; this condition required [Eq. (16) of Ref. 1]

$$Q(\epsilon) = \nu(\epsilon) P(\epsilon)/\langle \nu \rangle \tag{19}$$

where the average $\langle \cdots \rangle$ of any function $f(\epsilon)$ is defined by

$$\langle f \rangle \equiv \int d\epsilon f(\epsilon) P(\epsilon). \tag{20}$$

It should be noted that we are using the notation of Brissaud and Frisch[1] rather than that of Hooper;[12] our function $P(\epsilon)$ was called $Q(\epsilon)$ by Hooper and it is not the same as Hooper's well known $P(\epsilon)$. From Eq. (18) it is clear that $W(\epsilon|\epsilon')$ must be an even function for both ϵ and ϵ'; thus, in the general solution of Eq. (17),

$$\begin{aligned} P(\epsilon t|\epsilon_0 0) = {} & e^{-\nu(\epsilon)t}\delta(\epsilon-\epsilon_0) + \int_0^t dt_0\, e^{-\nu(\epsilon)(t-t_0)} W(\epsilon|\epsilon_0)\, e^{-\nu(\epsilon_0)t_0} \\ & + \sum_{n=1}^{\infty} \int_0^t dt_n \cdots \int_0^{t_1} dt_0 \int d\epsilon_n \cdots \int d\epsilon_1 \\ & \times e^{-\nu(\epsilon)(t-t_n)} W(\epsilon|\epsilon_n)\, e^{-\nu(\epsilon_n)(t_n-t_{n-1})} W(\epsilon_n|\epsilon_{n-1}) \cdots e^{-\nu(\epsilon_1)(t_1-t_0)} W(\epsilon_1|\epsilon_0)\, e^{-\nu(\epsilon_0)t_0}, \end{aligned} \tag{21}$$

it is clear that all terms in the sum over n are even, consequently the covariance takes a particularly simple form

$$\langle \boldsymbol{\epsilon}(t) \cdot \boldsymbol{\epsilon}(0) \rangle = \int d\epsilon \int d\boldsymbol{\epsilon}_0 \boldsymbol{\epsilon} \cdot \boldsymbol{\epsilon}_0 P(\epsilon t|\epsilon_0 0) P(\epsilon_0) = \int d\epsilon \epsilon^2 e^{-\nu(\epsilon)t} P(\epsilon). \tag{22}$$

Brissaud and Frisch then choose their function $\nu(\epsilon)$ so that Eq. (22) is equal to the electric field covariance which is well known from plasma kinetic theory.[13]

A major advantage of the model defined by Eqs. (18) and (19) is that it produces a simple expression for the average of the time development operator generated by the interaction $V(t) = \mathbf{M} \cdot \boldsymbol{\epsilon}(t)$, i.e.

$$i\partial_t T(t) = V(t) = \mathbf{M} \cdot \boldsymbol{\epsilon}(t) T(t). \tag{23}$$

For hydrogen lines, one readily obtains [Eq. (17) of Ref. 1]

$$\begin{aligned} I(\omega) \equiv \int e^{i\omega t} \langle T(t) \rangle\, dt = Re \Bigg[& \left\langle \frac{1}{\nu - i\omega + i\mathbf{M} \cdot \boldsymbol{\epsilon}} \right\rangle + \left\langle \frac{\nu}{\nu - i\omega + i\mathbf{M} \cdot \boldsymbol{\epsilon}} \right\rangle \\ & \times \left\langle \nu - \left(\frac{\nu^2}{\nu - i\omega + i\mathbf{M} \cdot \boldsymbol{\epsilon}} \right) \right\rangle^{-1} \left\langle \frac{\nu}{\nu - i\omega + i\mathbf{M} \cdot \boldsymbol{\epsilon}} \right\rangle \Bigg], \end{aligned} \tag{24}$$

which permits rather simple numerical calculations.

In order to examine more closely the validity of the Brissaud–Frisch model we first note that, in the static limit $\nu(\epsilon) \to 0$, Eq. (24) gives the well known static result

$$I(\omega) = \left\langle \frac{i}{\omega - \mathbf{M} \cdot \boldsymbol{\epsilon}} \right\rangle = i \int \frac{P(\epsilon)}{\omega - \mathbf{M} \cdot \boldsymbol{\epsilon}} d\boldsymbol{\epsilon}. \tag{25}$$

The limit $\nu(\epsilon) \to 0$ is a mathematical statement that the time between jumps in the electric field, $1/\nu(\epsilon)$, approaches infinity; that is, the field never changes. In the Brissaud–Frisch model, $\nu(\epsilon)$ is proportional to the ion plasma frequency, hence the static limit may be obtained by letting the ion mass approach infinity. The static result, Eq. (25), is also obtained in the far line wings $\omega \gg \mathbf{M} \cdot \boldsymbol{\epsilon}_0$ using $\mathbf{M} \cdot \boldsymbol{\epsilon}_0 > \nu(\epsilon_0)$, where ϵ_0 is the most probable ion field strength; these two inequalities are equivalent to those in Eq. (2a). To show this, we first note that the averages in Eq. (24) are essentially integrals of functions proportional to $P(\epsilon)/[\nu(\epsilon) - i(\omega - \mathbf{M} \cdot \boldsymbol{\epsilon})]$ which has two peaks, one at $\epsilon = \epsilon_0$ due to $P(\epsilon)$ and another at $\epsilon = \epsilon_1$ where $\epsilon = \mathbf{M} \cdot \boldsymbol{\epsilon}_1$. Since $\nu(\epsilon_0) \cong \omega_p$, the region of the integral around $\epsilon \cong \epsilon_0$ contributes a term which is proportional to (ω_p/ω) and is thus negligible in the far wings, $\omega \gg \mathbf{M} \cdot \boldsymbol{\epsilon}_0 > \nu(\epsilon_0) \cong \omega_p$. The integrand in the region $\epsilon \cong \epsilon_1$ is sharply peaked since both ω and $\mathbf{M} \cdot \boldsymbol{\epsilon}$ are much greater that $\nu(\epsilon_1)$; hence it may

be evaluated by a stationary phase integration. The first term on the right side of Eq. (24) thus reduces to the static result and the second term is of order $(\omega_p/\mathbf{M}\cdot\boldsymbol{\epsilon}_0)$ and thus negligible.

To study the dynamic limit, we use the result derived in Appendix A, where it is shown that $I(\omega)$ can be written in the form

$$I(\omega) = [-i\omega - M_c(\omega)]^{-1} \tag{26}$$

with M_c defined by

$$\begin{aligned} M_c(\omega) = &\int_0^\infty dt\, e^{i\omega t}\langle \mathbf{M}\cdot\boldsymbol{\epsilon}(t)\mathbf{M}\cdot\boldsymbol{\epsilon}(0)\rangle \\ &+ \int_0^\infty dt\, e^{i\omega t}\int_0^t ds\int_0^s ds'[\langle \mathbf{M}\cdot\boldsymbol{\epsilon}(t)\mathbf{M}\cdot\boldsymbol{\epsilon}(s)\mathbf{M}\cdot\boldsymbol{\epsilon}(s')\mathbf{M}\cdot\boldsymbol{\epsilon}(0)\rangle \\ &- \langle \mathbf{M}\cdot\boldsymbol{\epsilon}(t)\mathbf{M}\cdot\boldsymbol{\epsilon}(s)\rangle\langle \mathbf{M}\cdot\boldsymbol{\epsilon}(s')\mathbf{M}\cdot\boldsymbol{\epsilon}(0)\rangle] + \cdots \end{aligned}$$

This is the general result derived by Fano[14] in which each term will be evaluated using the probability functions $P(\boldsymbol{\epsilon}t|\boldsymbol{\epsilon}_0 0)$ obtained from Eqs. (18) and (21).

It is not surprising that the line shape, obtained by the model microfield theory, Eq. (24), can be written in the form obtained by Fano, Eqs. (26) and (27), because the latter are purely formal results which are independent of the model used for the actual calculations. That is, the Brissaud–Frisch MMM calculations using Eq. (24) are identical to the results of a calculation using Eq. (26) provided that every term in the series expansion of $M_c(\omega)$ is evaluated using the Brissaud–Frisch model. This fact permits a very convenient and informative analysis of the validity of the Brissaud–Frisch model in the dynamic limit.

4. THE MMM FOR BROADENING OF HYDROGEN LINES BY ELECTRONS

It has been known for sometime that, for hydrogen lines, the second order term in $M_c(\omega)$ produces virtually all of the dynamic aspects of electron Stark broadening. In fact, this was the only term considered in the early impact theories,[5] which used an impact parameter cutoff, b_0, to remove those interactions for which $\int_0^\infty V(t)\,dt > 1$; the contribution of these strong collisions was usually rather small and was approximated by a collision cross section πb_0^2. A more recent modified impact theory[15] used a frequency-dependent Lewis cutoff[16] which greatly improved the calculations for larger frequencies (i.e. frequency separations from line center $\Delta\omega$ greater than the electron plasma frequency ω_p) and an attempt was made to patch on an asymptotic line wing expression for the electrons.[17] Finally, calculations were made with a unified theory[7] which included some of the higher order terms in Eq. (27), namely, those corresponding to nonoverlapping binary collisions. In this theory, $M_c(\omega)$ was expressed as the Fourier transform of a function $F(t)$ and the $t\to 0$ and $t\to\infty$ asymptotes $F_0(t)$ and $F_\infty(t)$ were evaluated and analyzed. For values of ω near line center, it was found that $M_c(\omega)$ is determined primarily by $F_\infty(t)$ and this gave results essentially identical to the second order results of the modified impact theory.[15] In the line wings, $M_c(\omega)$ is determined by $F_0(t)$ and one obtains the familiar Holtsmark asymptotic result in which correlations between electrons are neglected; it is known that this result is less accurate than a static calculation which includes electron correlation effects, but it was argued that this small error would be overwhelmed by the large contribution from static ion broadening. Thus, while the unified theory improved the theoretical foundations for electron broadening and to some extent the accuracy and reliability of numerical calculations, this theory also tended to confirm the original idea that, for electron broadening at least, one needs only the electric field autocorrelation function (the second order term in $M_c(\omega)$) in the dynamic limit and a static asymptote for the line wings; any reasonably smooth means of joining these two asymptotes seems to produce only small errors in the transition region. Additional support for this conclusion comes from studies of time ordering,[18,19] electron correlations,[20] and other effects[21] which appear only in fourth and higher order terms in $M_c(\omega)$. These corrections[22] are always found to be at the 10% level thus confirming the idea that 90% or more of the electron broadening is described by the electric field autocorrelation and an appropriate static limit.

From our previous discussion of the kangaroo process (KP) proposed by Brissaud and Frisch, it is clear that their model gives the same second order results for $M_c(\omega)$ as the unified theory (recall that their jumping frequency $\nu(\epsilon)$ was defined in such a way that their calculation of the electric field autocorrelation, Eq. (2.15), would equal the result known from plasma kinetic theory).

A calculation of $M_c(\omega)$ with the Brissaud–Frisch model also includes all higher order terms; the KP model calculations have not actually been done this way, but as noted in the previous section and in Appendix A, the KP calculations can be rewritten in the form of Eq. (26), thus permitting an identification of each term in $M_c(\omega)$. Taking the fourth order term for example [Appendix A, Eq. (A.7)], we see that this involves a calculation of

$$M_4(\omega) = \int_0^\infty e^{i\omega t_2}\, dt_3 \int_0^{t_3} dt_2 \int_0^{t_2} dt_1\, C_4(t_3 t_2 t_1), \tag{28}$$

$$C(t_3 t_2 t_1) \equiv \langle V(t_3) V(t_2) V(t_1) V(0)\rangle - \langle V(t_3) V(t_2)\rangle \langle V(t_1) V(0)\rangle. \tag{29}$$

In the KP model, these averages are evaluated using the approximation specified by Eq. (18) to simplify the general Markovian probability function $P(\epsilon t|\epsilon' t')$ given in Eq. (21). In the unified theory, only the term $\langle V(t_3) V(t_2) V(t_1) V(0)\rangle$ is evaluated (the other term comes from the projection operator which is neglected in the unified theory calculations; see Appendix B of Ref. 18 for the justification of this approximation); it is also assumed that all four functions $V(t)$ represent one and the same electron (i.e. overlapping collisions are neglected) and, finally, the time ordering $t_3 \geq t_2 \geq t_1 \geq 0$ in Eq. (28) is usually neglected. Thus, we see that with the unified theory one calculates a highly restricted subset of the higher order terms using various approximations (e.g. binary collisions, neglecting time ordering, etc.) whereas with the MMM one calculates all of the higher order terms using the function $P(\epsilon t|\epsilon' t')$. The accuracy of any MMM calculation for these higher order terms will depend on the particular model chosen for $P(\epsilon t|\epsilon' t')$. We emphasize that the validity of the MMM for higher order dynamic effects, such as time ordering is really a question of its validity for the higher order terms in $M_c(\omega)$ in the dynamic limit; in principle, all of these terms exist in an MMM calculation but their accuracy will depend on the model chosen for $P(\epsilon t|\epsilon' t')$.

Broadening by electrons is not likely to provide a sensitive test for these higher order terms because, as noted above, they produce only a 10% change in the line profile. In fact, Voslamber[23] has compared a KP model calculation by Seidel[24] with his unified theory calculations and finds at most a 5% difference between the two profiles (i.e. a 10% difference in the electron contribution) in the near line wings (i.e. for frequencies between the plasma frequency and the Weisskopf frequency).

One important result of the inclusion of all higher order terms in $M_c(\omega)$ is that the MMM approaches the correct many-body static asymptote, whereas the unified theory approaches the on-particle static asymptote known as the Holtsmark limit. This may be expected to produce a difference of 5% or less for frequencies around the Weisskopf frequency (for much larger frequencies, the many-body static results approach the Holtsmark limit and, for much smaller frequencies, static effects vanish). In fact, part of the discrepancy observed by Voslamber[23] may have been due to this difference in the calculation of static effects.

The preceding discussion may be summarized by the following points: (1) the MMM is in principle more powerful than the unified theory because it provides a calculation of all higher order terms in $M_c(\omega)$ rather than a restricted subset of terms; (2) in the static limit this enables the MMM to use the best known many-body probability distribution[12] rather than the one-particle Holtsmark limit used in the unified theory; (3) in the dynamic limit, the accuracy of an MMM calculation of the higher order terms (which include time ordering effects, electron correlations, etc.) will depend on the particular model chosen for $P(\epsilon t|\epsilon' t')$; (4) dynamic electron broadening is determined almost entirely by the second order term in $M_c(\omega)$ (essentially the electric field autocorrelation function) hence one may expect very good results from the Brissaud–Frisch KP model because this model, by its construction, gives the known second order results; (5) this insensitivity to higher order dynamic effects means that one will not be able to use electron broadening to distinguish between the Brissaud–Frisch model and various alternative stochastic models.[25]

We may thus conclude that the KP model proposed by Brissaud and Frisch provides a powerful theory for electron broadening of hydrogen lines; it is as accurate as the unified theory to second order and it is more accurate in the static limit. The validity of the KP model for higher order dynamic effects has not been established as yet because these effects make only a 10% contribution to electron broadening.

5. THE MMM FOR BROADENING OF HYDROGEN LINES BY IONS

For many years, ions were treated as static for all line broadening calculations. It was known that ion dynamic effects could be present at line center but it was thought that they would not produce an observable change in the line profile. However, measurements[26,27] of the halfwidth of H_α and the central dip of H_β for hydrogen and deuterium perturbed by various noble gases showed a $1/\sqrt{\mu}$ dependence on the reduced mass μ and agreement was obtained with static ion calculations only in the limit $\mu = \infty$. Furthermore, a recent measurement[28] of the Ly_α halfwidth is 2.5 times larger than that calculated using a static ion approximation.

Measurements such as these have stimulated many attempts to use the impact and unified theories to describe the dynamic ion broadening. Such attempts have met with very little success because both of these theories are based on the fundamental assumption that the radiating atom is perturbed by a series of nonoverlapping binary collisions with the perturbers whereas, for densities of 10^{17} or higher, the ions are characterized by important overlap effects as discussed in Section 2. These overlap effects are seen in the static limit where the many body probability distribution[12] must be used rather than the one particle Holtsmark distribution which is the normal static limit of the unified theory. Attempts have been made to modify the unified theory by building in the many body static limit but this does not address the problem of overlap in the dynamic limit.

Since the MMM does not employ by a binary collision assumption, it was hoped that this type of theory would provide a good theoretical treatment of ion broadening including both the many body static limit as well as an improved treatment of dynamic effects. Seidel[4] has performed detailed MMM calculations for H_α, H_β and Ly_α using the Brissaud–Frisch KP model for both ions and electrons and his calculations give much better agreement with experimental data than previous calculations using a purely static theory for the ions but some important discrepancies remain, particularly for the H_β dip at lower densities (Fig. 9 of Ref. 4). These results indicate that the ability of the MMM to include ion dynamic effects constitutes a major improvement over previous theories but something is still missing in the calculations. Seidel[29] has investigated the influence of radiator motion on the collision broadening but it was found that this effect is negligible for the hydrogen lines considered.[30] The fact that something is still missing in Seidel's MMM calculations has led us to reexamine the validity of the Brissaud–Frisch model as applied to ions.

In the previous section, it was noted that the Brissaud–Frisch KP model gave good results for electron broadening because the known electric field autocorrelation and many body static probability functions were used to define certain parameters in their model. This insured that their line profile would be correct in the static limit and it would also give the correct second order approximation for $M_c(\omega)$ which represents about 90% of the electron dynamic effects. This second order approximation was sufficient for the electrons because the average electron–atom collision is weak. However, the average ion-atom collision is strong so one would expect the higher order terms in $M_c(\omega)$ to be important and the validity of the KP model has not yet been established for these terms in the dynamic limit (i.e. values of ω near line center).

Before proceeding with a further examination of the KP model, it may be helpful to briefly outline some of the results obtained from the study of rotational and vibrational linewidths of molecules in electrically neutral gases; for these systems virtually all collisions are strong hence they can provide some insight into the theoretical requirements for broadening by strong dynamic collisions. For many years, it was thought that one could use a second order approximation for the averaged binary collision S matrix; strong collisions were treated by introducing an impact cutoff at some value b_0 and adding a strong collision cross section πb_0^2 just as in the early impact theories for electrons. The value of b_0 should be approximately equal to the Weisskopf radius r_w defined in Eq. (2b), however, for molecules πr_w^2 is responsible for a large part of the observed halfwidth and in many cases this "correction" by itself gave

halfwidths which were larger than the experimentally observed values. Consequently many heuristic adjustments of the impact parameter cutoff were needed in order to produce agreement with experimental data, often resulting in ridiculous values for b_0 which were substantially smaller than the hard sphere diameter for the particles;[31] it was finally concluded that this approach was completely invalid. This conclusion has never been reached in the field of Stark broadening simply because πb_0^2 constitutes a relatively small correction to the electron halfwidth and it was not necessary to study the strong collision problem in any detail. The whole cutoff procedure is based on an argument that strong collisions, $b < b_0$, simply cause the S matrix to oscillate rapidly between +1 and 1 and, under the impact parameter average, $\int S(b) b \, db$, the contribution from $0 \leq b \leq b_0$ is essentially zero; since $M_c(0)$ is proportional to the average of $2\pi(1 - S)$, the integral of 1 over $(0, b_0)$ produces the strong collision cross section πb_0^2. In recent years, a molecular line broadening theory has been developed[32] in which S is approximated by $\exp(-i\eta)$ where η is a matrix defined in such a way that the approximate S matrix is rigorously correct to second order in the interaction and the errors introduced in higher order terms are small (10–15%) and tend to cancel one another when all terms in the series expansion are added together. This theory was found to give very good agreement with experimental data[32,33] without any recourse to cutoffs or other adjustable parameters. Using this theory, it was also found[34] that the S matrix does indeed oscillate for small impact parameters but this oscillation is not a rapid and uniform function of b hence S does not integrate to zero for small impact parameters. It was thus established that the arguments based on rapid oscillation of the S matrix are false and the strong collision cutoff procedure provides a very poor description of strong dynamic collisions.

From the experience gained in molecular line broadening, it would seem that we want an approximation for $M_c(\omega)$ which is rigorously correct to second order and which gives a reasonable approximation for each of the higher order terms. The unified theory is certainly correct to second order but the higher order terms will not be valid for ions, even if time ordering is included, because overlap effects are neglected. The MMM proposed by Brissaud and Frisch is also correct to second order and, as noted already, all terms in the expansion of $M_c(\omega)$ are included; it only remains to examine the accuracy of their KP model for these higher order terms.

We must note at this point that Brissaud *et al.*[9] have examined the validity of the KP and they have briefly discussed its treatment of strong interactions [see Eq. (3.19) of Ref. 9]. Unfortunately, they have assumed that all strong fields are static (see Sect. 2.1 of Ref. 9) which is not correct, as already noted in Section 2 of the present paper; they also assumed that a strong field is produced by a single particle [Eq. (2.3) of Ref. 9] thereby neglecting the overlap of strong binary collisions which is important for ion fields. In their treatment of strong collisions, Section 3.3 of Ref. 9), they assert, without proof, that the effect of strong fields is to instantaneously damp the correlation function $\langle T(t) \rangle$ and their damping rate, $1/\Delta t$, is just the usual damping rate obtained from the strong collision cross section πb_0^2. Their result, Eq. (3.19) of Ref. 19, is thus equivalent to the usual strong collision cutoff procedure because multiplying $\langle T(t) \rangle$ by a damping rate $\exp(-\gamma t)$ is equivalent to adding γ, the strong collision cutoff corrections, to the halfwidth. Brissaud *et al.* obviously thought that this result was desirable because it is listed on p. 1142 of Ref. 9 in items (iii) and (iv) which they put forth as the strong points of the KP model; however, as noted above, the analysis of systems which are dominated by strong collisions has shown that this strong collision cutoff procedure is seriously in error. On the other hand, the analysis presented in Ref. 9 is clearly incorrect because it was assumed that (1) all strong fields are static, (2) each strong field is due to only one particle, and (3) strong fields simply serve to damp $\langle T(t) \rangle$. None of these assumptions is needed in the derivation of the Brissaud–Frisch KP model;[1,2] hence, one may still hope that the KP provides an adequate treatment of strong collisions even though the results obtained in Ref. 9 would indicate that it is no better than an impact parameter cutoff procedure.

To gain further insight into the physical nature of the KP model and its suitability for ion fields, we next consider the validity of the product form, Eq. (18), which is used for the transition rate $W(\epsilon|\epsilon')$. The physical meaning of W may be obtained from Eq. (12) which says that, if the electric field takes the value ϵ' at some instant of time, $\Delta t W(\epsilon|\epsilon')$ is, in the limit $\Delta t \to 0$, the probability that the electric field will jump to the value ϵ during the time interval Δt.

The product form used by the KP model, $W(\epsilon|\epsilon') = Q(\epsilon)\nu(\epsilon')$, says that, if the electric field takes the value ϵ' at some instant, $\Delta t\nu(\epsilon')$ is probability that there will be a jump to some other value during the time interval Δt and the probability of finding the value ϵ after the jump is $Q(\epsilon)$. The fact that $\Delta t\ W(\epsilon|\epsilon')$ is a product of these two probabilities, namely, $\Delta t\nu(\epsilon')$ and $Q(\epsilon)$, means that the two events are completely independent; that is, the value of the electric field after a jump is completely independent of the value it had before the jump. Thus, the KP is said to be a Markov process *with no memory*. For the sake of comparison, suppose we consider a model in which

$$W(\epsilon|\epsilon') = f(\epsilon - \epsilon')\nu(\epsilon'), \tag{30}$$

$$\int d\epsilon f(\epsilon - \epsilon') = 1, \tag{31}$$

and $f(\delta\epsilon)$ is peaked about $\delta\epsilon = 0$. In this model, $\Delta t\nu(\epsilon')$ would again be the probability that the electric field of strength ϵ' will make a jump during the time Δt but the probability $f(\epsilon - \epsilon')$ of finding the value ϵ after the jump is peaked around the value ϵ' before the jump. This is an example of a Markovian model *with memory*. To compare these two kinds of Markovian models we consider the variation in the electric field produced by a typical binary collision; at the beginning of the collision the electric field will increase from some background level up to a maximum value at the time of closest approach, then it will decrease to the fluctuating background level again. The goal of any MMM is to represent such a collision, as well as other overlapping collisions etc., with a stepwise continuous electric field which has the same general behavior; that is, it increases in several discrete jumps to a maximum value and then decreases back to a fluctuating background level. The average duration and intensity of such events will be determined by the statistics of the model chosen and the criterion for success will be the ability of the model to predict the correlation functions needed for line profile calculations. In the KP model, the value of the electric field after a jump is completely independent of the value before the jump, hence a large positive value ϵ' may be followed by another large positive value ϵ which is near ϵ' but it is just as likely to be followed by a small value ϵ because $Q(\epsilon)$ is peaked near small not large values of ϵ. It is hard to imagine how such a model could ever represent the type of electric field variations present in a plasma. In the Markovian model with memory, Eqs. (30) and (31), the fact that f is peaked about zero insures that a large value ϵ' will be followed by a value ϵ near ϵ' hence we would expect this type of model to be capable of representing the plasma electric field fluctuations.

At this point, one may wonder how the KP model could ever have any success in representing the plasma electric field. It must therefore be recalled the KP model has two "adjustable parameters" $Q(\epsilon)$ and $\nu(\epsilon)$ which were defined in such a way that the static probability distribution and the electric field autocorrelation function would be equal to the known values. Since these are essentially the only functions needed for electron broadening, the KP was satisfactory. On the other hand, since higher order terms in $M_c(\omega)$, that is, higher order electric field correlation functions, are required for dynamic ion broadening effects, it is no longer surprising that the KP is incapable of calculating these functions.

From these arguments, we conclude that the KP may be unsuitable for ion broadening because it does not provide a good representation of the plasma electric field fluctuations. This model was successful for electron broadening only because the latter is determined primarily by the static limit and the electric field autocorrelation function and it was possible to adjust the free parameters in the KP to insure that these two functions would be correct. It also seems that the KP will not be capable of improving theoretical calculations in the transition region between the line center and the line wings (i.e. between the plasma frequency and the Weisskopf frequency) where higher order terms in $M_c(\omega)$ become important and where 10% uncertainties still exist in unified theory calculations. On the other hand, it seems that a Markovian model with memory, such as Eqs. (30) and (31), might give a better representation of the physical processes in a plasma and might, therefore, be used for an improved MMM.

6. THE MMM FOR BROADENING OF ISOLATED HELIUM LINES

For a hydrogen atom, the energy levels characterized by different values of the orbital quantum number l are effectively degenerate (within the manifold of each principle quantum

number n). This means that the dipole operator M will have nonvanishing matrix elements between degenerates states and the lowest order interaction with an electric field is $\mathbf{M} \cdot \boldsymbol{\epsilon}$. For helium atoms, the states corresponding to different l values are split by $L \cdot S$ coupling and M has no nonvanishing matrix elements between degenerate states; in this case for isolated lines (i.e. nondegenerate), the lowest order interaction with the electric field is the second order result $\Sigma_k \mathbf{M} \cdot \boldsymbol{\epsilon}|k\rangle (E_k - H_0)^{-1} \langle k|\mathbf{M} \cdot \boldsymbol{\epsilon}$ where $|k\rangle$ and E_K denote the eigenvectors and eigenvalues of H_0. That is, for isolated lines in helium the interaction is proportional to ϵ^2 or $1/r^4$ whereas for hydrogen the interaction in proportional to ϵ or $1/r^2$. This means that, by comparison with hydrogen line broadening, the helium lines are broadened by short range interactions so there is less broadening due to long range weak interactions and there is a bigger contribution from strong interactions (e.g. the "strong collision correction" resulting from the strong collision cutoff procedure is typically 20–80% for helium lines[35] compared with typically 10% for hydrogen lines). For ions we may expect a similar stress on strong short range dynamic effects. Thus, if the KP is unsuitable for the higher order terms in $M_c(\omega)$, as argued in section 5, we would expect it to give less satisfactory results for helium lines than for hydrogen lines. This is in fact the case for measurements performed on the forbidden lines at 4922 and 4471 Å, as evidenced by the results of Mazure *et al.*[36] and unpublished results reported by Helbig.[37] This relatively poor treatment of ion dynamics for helium thus supports our argument that the KP is not adequate for the higher order dynamic terms required for a description of strong dynamic interactions.

7. CONCLUSIONS

From the analysis of Markovian model microfield methods presented in this paper, we draw the following conclusions;

(1) The MMM provides a means of calculating all terms in the operator $M_c(\omega)$ obtained by Fano[14] whereas the unified theory[6,7] calculates only the subset of terms which correspond to nonoverlapping binary collisions for statistically independent perturbers.

(2) This means that the MMM includes those higher order terms which represent the effects known as time ordering, overlap of strong collisions, higher order correlations, etc. The validity of the MMM calculation of these higher order terms will depend on the specific model chosen for the conditional probability $P(\boldsymbol{\epsilon}t|\boldsymbol{\epsilon}'t')$ and the transition rate $W(\boldsymbol{\epsilon}|\boldsymbol{\epsilon}')$ discussed in Section (3).

(3) Throughout this paper we make a distinction between the model microfield method (MMM) in general, which may refer to any stochastic model for $\boldsymbol{\epsilon}(t)$, and the kangaroo process (KP) which is a specific type of MMM. The KP model, proposed by Brissaud and Frisch[1,2] and used by Seidel,[3,4] is defined by the choice $W(\boldsymbol{\epsilon}|\boldsymbol{\epsilon}') = Q(\epsilon)\nu(\epsilon')$ with $\nu(\epsilon')$ and $Q(\epsilon)$ being defined in terms of the known electric field autocorrelation function $\langle \boldsymbol{\epsilon}(t) \cdot \boldsymbol{\epsilon}(0)\rangle$ and the static electric field distribution function $P(\epsilon)$ [see Eqs. (19) and (22)].

(4) The KP model gives good results for electron broadening of hydrogen lines because the choice of ν and Q guarantees that $M_c(\omega)$ will be correct to second order and the static limit will be correct; for electron broadening, these two limits determine 90% or more of the value of the intensity for any frequency of interest.

(5) For ion broadening of hydrogen lines the KP gives better results than the unified theory but certain obvious deficiencies remain in the density dependence of the dip in H_β etc.[4] In Section 5, it was argued that a Markovian model without memory, such as the KP, actually gives a very poor representation of the electric field fluctuations in a plasma and, for this reason, the KP model is not capable of correctly treating the higher order $M_c(\omega)$ terms which are important for ion broadening. That is, the unified theory completely ignores the higher order correlation and collision overlap terms and thus gives poor results for ion dynamic effects. The KP includes these terms but calculates them with an inadequate model for $P(\boldsymbol{\epsilon}t|\boldsymbol{\epsilon}'t')$ thereby giving improved results; nonetheless, obvious errors remain due to the unsuitability of the KP model for the higher order terms which are required for strong dynamic interactions.

(6) Our argument that the KP model is unsuitable for higher order dynamic terms is supported by the fact that KP model calculations give relatively poor results for helium lines (i.e. compared to the KP results for hydrogen lines). We argue, in Section 6, that helium lines are more sensitive to strong dynamic interactions due to the relatively short range interaction for helium.

(7) In Section 5 it was shown that a Markovian model with memory, such as that defined by Eqs. (30) and (31), can give a better representation of the physical processes in the plasma (i.e. the electric field fluctuations) and should therefore give a better treatment of ion broadening.

(8) Although we have discussed only Markovian models for simplicity, one could equally well construct an MMM based on a nonMarkovian model which could include more memory effects than those represented by Eqs. (30) and (31). At the present time there is no reason to think that a non-Markovian model would be inherently superior because the known non-Markovian properties of line broadening (e.g. Section 3.3) of Ref. 14) result not from the electric field statistics but rather from the fact that one has derived an equation of motion for a subsystem, the radiator, by averaging over another subsystem, the perturbers (e.g. Section 2.B of Ref. 7). That is, the same non-Markovian line broadening equation will result whether we choose Markovian or non-Markovian statistics for the perturber subsystem.

(9) It may be useful to summarize the Markovian model microfield approach by noting that it involves three levels of approximations. First, there is the fundamental assumption that the true statistics of the electric microfield are adequately represented by a Markov model as defined by Eq. (8); the validity of this assumption has not been studied as yet. Next, there is the choice of a specific Markovian model such as the KP proposed by Brissaud and Frisch or the model with memory specified by Eqs. (30) and (31); in fact, there are many different Markov models[38] one can choose from. The choice of the model is of course dictated by the statistical properties one wants to represent; for example, the KP model gives the correct electric field autocorrelation but it seems to fail for the higher order correlation functions, etc. Finally, it may be necessary to make additional approximations in order to obtain a soluable line shape expression. This is not necessary when using the KP model since that model produces a calculable line shape expression with no further approximations; however, improved Markov models, such as that proposed in Eqs. (30) and (31), will probably not permit actual line profile calculations without further simplifying approximations and the validity of these additional approximations must also be considered when comparing various alternative Markov models.

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APPENDIX

In this appendix, we present the algebra necessary to cast the MMM line shape expression, Eq. (24), in the form obtained by Fano, Eq. (26), which is familiar to users of the unified and impact theories. From Eq. (24), we obtain

$$\begin{aligned} I(\omega) &= \left\langle \frac{1}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \left[1 + \nu \left\langle \frac{\nu(-i\omega + \mathbf{M}\cdot\boldsymbol{\epsilon})}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle^{-1} \left\langle \frac{\nu}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \right] \right\rangle \\ &= \left\langle \frac{1}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \left[1 + \frac{\nu}{-i\omega + \alpha(\omega)} \right] \right\rangle \\ &= \left\langle \frac{\nu - i\omega + \alpha(\omega)}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \left(\frac{1}{-i\omega + \alpha(\omega)} \right) \\ &= \frac{1 + \beta(\omega)}{-i\omega + \alpha(\omega)} \\ &= \frac{1}{-i\omega + M_c(\omega)}, \end{aligned} \tag{A1}$$

where

$$\alpha(\omega) \equiv \left\langle \frac{\nu(i\mathbf{M}\cdot\boldsymbol{\epsilon})}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \left\langle \frac{\nu}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle^{-1} \tag{A2}$$

$$\beta(\omega) \equiv \left\langle \frac{\alpha(\omega) - i\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \tag{A3}$$

$$M_c(\omega) \equiv i\omega + [-i\omega + \alpha(\omega)]/[1 + \beta(\omega)] = [\alpha(\omega) + i\omega\beta(\omega)]/[1 + \beta(\omega)]. \tag{A4}$$

Using $\langle \mathbf{M}\cdot\boldsymbol{\epsilon} \rangle = 0$, we obtain to lowest order

$$\begin{aligned} \alpha(\omega) + i\omega\beta(\omega) &= \left\langle \frac{\nu(i\mathbf{M}\cdot\boldsymbol{\epsilon})}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \left\langle \frac{\nu}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle^{-1} \left[1 + i\omega \left\langle \frac{1}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \right] - i\omega \left\langle \frac{i\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \\ &= \left\langle \frac{\nu(i\mathbf{M}\cdot\boldsymbol{\epsilon})}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle + \left\langle \frac{\nu(i\mathbf{M}\cdot\boldsymbol{\epsilon})}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \left\langle \frac{\nu}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle^{-1} \left\langle \frac{i\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle - i\omega \left\langle \frac{i\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \\ &= \left\langle (i\mathbf{M}\cdot\boldsymbol{\epsilon}) + \frac{(\mathbf{M}\cdot\boldsymbol{\epsilon})^2}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle + \left\langle \frac{\nu(i\mathbf{M}\cdot\boldsymbol{\epsilon})}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \left\langle \frac{\nu}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle^{-1} \left\langle \frac{i\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega + i\mathbf{M}\cdot\boldsymbol{\epsilon}} \right\rangle \\ &= \left\langle \frac{(\mathbf{M}\cdot\boldsymbol{\epsilon})^2}{\nu - i\omega} \right\rangle - \left\langle \frac{(\mathbf{M}\cdot\boldsymbol{\epsilon})^4}{(\nu - i\omega)^3} \right\rangle + \left\langle \frac{\nu}{\nu - i\omega} \right\rangle^{-1} \left\langle \nu \left(\frac{\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega} \right)^2 \right\rangle \left\langle \left(\frac{\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega} \right)^2 \right\rangle + \cdots \end{aligned} \tag{A5}$$

$$\beta(\omega) = \left\langle \nu \left(\frac{\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega} \right)^2 \right\rangle \left\langle \frac{\nu}{\nu - i\omega} \right\rangle^{-1} \left\langle \frac{1}{\nu - i\omega} \right\rangle - \left\langle \left(\frac{\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega} \right)^2 \right\rangle + \cdots; \tag{A6}$$

thus

$$\begin{aligned} M_c(\omega) &= \left\langle \frac{(\mathbf{M}\cdot\boldsymbol{\epsilon})^2}{\nu - i\omega} \right\rangle - \left\langle \frac{(\mathbf{M}\cdot\boldsymbol{\epsilon})^4}{(\nu - i\omega)^3} \right\rangle + \left\langle \frac{\nu}{\nu - i\omega} \right\rangle^{-1} \left\langle \nu \left(\frac{\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega} \right)^2 \right\rangle \left\langle \left(\frac{\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega} \right)^2 \right\rangle + \cdots \\ &\quad - \left\langle \frac{(\mathbf{M}\cdot\boldsymbol{\epsilon})^2}{\nu - i\omega} \right\rangle \left[\left\langle \nu \left(\frac{\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega} \right)^2 \right\rangle \left\langle \frac{\nu}{\nu - i\omega} \right\rangle^{-1} \left\langle \frac{1}{\nu - i\omega} \right\rangle - \left\langle \left(\frac{\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega} \right)^2 \right\rangle \right] + \cdots \\ &= \left\langle \frac{(\mathbf{M}\cdot\boldsymbol{\epsilon})^2}{\nu - i\omega} \right\rangle - \left\langle \frac{(\mathbf{M}\cdot\boldsymbol{\epsilon})^4}{(\nu - i\omega)^3} \right\rangle + \frac{1}{i\omega} \left\langle \frac{\nu}{\nu - i\omega} \right\rangle^{-1} \left\langle \nu \left(\frac{\mathbf{M}\cdot\boldsymbol{\epsilon}}{\nu - i\omega} \right)^2 \right\rangle - \frac{1}{i\omega} \left\langle \frac{(\mathbf{M}\cdot\boldsymbol{\epsilon})^2}{\nu - i\omega} \right\rangle^2 + \cdots \\ &= \int_0^\infty dt\, e^{-i\omega t} \left[\left\langle e^{-\nu t} (\mathbf{M}\cdot\boldsymbol{\epsilon})^2 \right\rangle - \int_0^t ds \int_0^s ds' \left\langle e^{-\nu(t+s+s')} (\mathbf{M}\cdot\boldsymbol{\epsilon})^4 \right\rangle \right] \\ &\quad + \frac{1}{i\omega} \int_0^\infty dt\, e^{-i\omega t} \int_0^t ds \langle \nu\, e^{-\nu(t+s)} (\mathbf{M}\cdot\boldsymbol{\epsilon})^2 \rangle \sum_1^\infty \frac{1}{\langle \nu \rangle^{n+1}} \left[\int_0^\infty e^{i\omega t} \langle \nu^2 e^{-\nu t} \rangle \right]^n \\ &\quad - \frac{1}{i\omega} \left[\int_0^\infty e^{-i\omega t} \cdot e^{-\nu t} (\mathbf{M}\cdot\boldsymbol{\epsilon})^2 \rangle \right]^2 + \cdots \end{aligned} \tag{A7}$$

$$= \int_0^\infty dt\, e^{-i\omega t}\langle v(t)v(0)\rangle$$

$$- \int_0^\infty dt\, e^{-i\omega t} \int_0^t ds \int_0^s ds' \int d\epsilon_1 \int d\epsilon_2 \int d\epsilon_3 \int d\epsilon_4 (\mathbf{M}\cdot\epsilon_1)(\mathbf{M}\cdot\epsilon_2)(\mathbf{M}\cdot\epsilon_3)(\mathbf{M}\cdot\epsilon_4)$$

$$P(\epsilon_1 t|\epsilon_2 s)P(\epsilon_2 s|\epsilon_3 s')P(\epsilon_3 s'|\epsilon_4 0)P(\epsilon_4)$$

$$-\frac{1}{i\omega}\left[\int_0^\infty e^{-i\omega t}\langle v(t)v(0)\rangle\right]^2 + \cdots$$

$$= \int_0^\infty e^{-i\omega t}\Big[\langle v(t)v(0)\rangle - \int_0^t ds \int_0^s ds'\langle v(t)v(s)v(s')v(0)\rangle$$

$$+ \int_0^t ds \int_0^s ds'\langle v(t)v(s)\rangle\langle v(s')v(0)\rangle\Big) + \cdots$$

where $v(t) = \mathbf{M}\cdot\epsilon(t)$ and in a Markovian MMM,

$$\langle v(t)v(s)v(s')v(0)\rangle = \int d\epsilon_1 \int d\epsilon_2 \int d\epsilon_3 \int d\epsilon_4 (\mathbf{M}\cdot\epsilon_1)(\mathbf{M}\cdot\epsilon_2)(\mathbf{M}\cdot\epsilon_3)(\mathbf{M}\cdot\epsilon_4)$$

$$\times P(\epsilon_1 t|\epsilon_2 s)P(\epsilon_2 s|\epsilon_1 s')P(\epsilon_3 s'|\epsilon_3 0)P(\epsilon_3). \tag{A8}$$

A term such as Eq. (A8) splits into a sum of two terms when using Eqs. (18) and (21), the first is a product of three delta functions giving rise to the $(\mathbf{M}\cdot\epsilon)^4$ term in Eq. (A7) and the second is a product of two delta functions, $\delta(\epsilon-\epsilon_2)\times\delta(\epsilon_3-\epsilon_4)$, which gives rise to the summation term in Eq. (A7); the terms with less than two delta functions vanish because $\langle\mathbf{M}\cdot\epsilon\rangle = 0$ or rather, ν and Q are even functions of ϵ whereas $\mathbf{M}\cdot\epsilon$ is odd.

Thus, we see that the MMM line shape can be written in the form derived by Fano where each term in the expansion of $M_c(\omega)$ is evaluated using the conditional probability $P(\epsilon t|\epsilon' t')$ as in Eq. (A8) or Eq. (22). Actually, one could have simply started with Fano's result since it is derived with no approximations (other than the statistical independence of radiator and perturber subsystems) and then, choosing an MMM, each term in $M_c(\omega)$ would be evaluated as in Eq. (A8).