Progresses in Monte Carlo simulation of light scattering spectra in atomic gases

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Abstract. The dynamics of the spontaneous fluctuations of a equilibrium system is related to the system response to small perturbations. It therefore, contains information on thermodynamic and transport properties. Numerical experiments based on the DSMC methods have been performed to calculate the spectrum of density and correlations in gaseous Argon [1]. In the kinetic regime, differences with the predictions of a BGK kinetic model are discussed. These results are relevant in view of the renewed interest in Rayleigh–Brillouin scattering in gases made possible by the use of nonlinear optical techniques [2-4]. New calculations based on a particle solution of the BGK equation [5] allow to clarify the relation between dynamics correlations produced by DSMC and the kinetic model.

Keywords: equilibrium fluctuations, particle simulation, BGK model.
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INTRODUCTION

The dynamics of the spontaneous fluctuations of equilibrium systems is related to the system response to small perturbations. It therefore contains information on thermodynamic and transport properties [6].

The dynamics of the fluctuations of a dynamic variable A(**r**,t) is discussed by introducing the space-time correlation function:

$$\delta A^2(\mathbf{r},t;\mathbf{r}',t') \equiv \left\langle \delta A(\mathbf{r}',t')\delta A(\mathbf{r},t)\right\rangle = \delta A^2(|\mathbf{r}-\mathbf{r}'|,t-t') \tag{1}$$

where $\langle ... \rangle$ means ensemble average, $\delta A(\mathbf{r},t) = A(\mathbf{r},t) - \langle A(\mathbf{r},t)\rangle$ is the fluctuation of the dynamic variable and the last equality comes from the isotropy and equilibrium assumptions. In particular, the quantity actually measured in light scattering experiments is the power spectrum of density fluctuations [6]:

$$\delta n^2(k,\omega) = \int e^{i(kr-\omega t)}\delta n^2(r,t)\,dr\,dt \tag{2}$$

The ratio of the fluctuation wavelength to the mean free path defines the flow regime (from high to low ratios: hydrodynamic, kinetic, collisionless). In light scattering experiments the probed wavelength is related to that of the incident light via the Bragg condition. Therefore, light scattering in gases mainly probes the response of the system in the kinetic regime. Analytical theories are only available for the two opposite cases of completely collisionless fluids [7] and fluids described by the Navier-Stockes (NS) equations [8, 9]. Approximate theories based on the BGK collision term have been derived for the kinetic regime [9, 10]. However, it is known that the BGK equation, in the hydrodynamic limit gives [11]:

$$\mathrm{Pr} \equiv \frac{\mu c_p}{\lambda} = 1 \tag{3}$$

where μ is the shear viscosity of the gas, c_p the constant pressure specific heat and λ the thermal conductivity. The BGK model parameter can therefore be adjusted in order to match the real thermal conductivity or the viscosity, but not both.

The use of Monte Carlo particle methods for the study of hydrodynamic fluctuations in gases has been demonstrated many years ago by M. Malek Mansour and coworkers [12] who studied a nonequilibrium gas subject to a fixed heat flux; analogous results were recently found by us for the equilibrium fluctuations in the hydrodynamic regime [13]. The study was then extended to the complete range of regimes, from collisionless to hydrodynamic [1]. All these works employed Direct Simulation Monte Carlo (DSMC) [14] as the simulation tool. Recent advances in the GRID computing infrastructure [15] allow very fine details to be resolved at sufficient accuracy to distinguish between the predictions of different theoretical models, which could not be possible in the framework of previous numerical capabilities.

In particular, in this work we focus on the simulation of light scattering spectra in the kinetic regime and on the differences that arise with the use of different kinetic models. We compare the results of DSMC simulations, the results of particle simulation of the full BGK equation [5] and the results of a linearised BGK model [10].
These results are also relevant in view of the renewed interest in Rayleigh-Brillouin scattering in gases made possible by the use of nonlinear optical techniques [2-4]. Coherent Rayleigh-Brillouin scattering is a technique capable of making localized and high signal-to-noise ratio measurements of gases from the collisionless limit to the hydrodynamic regime. CRBS data are therefore expected to become a valuable source for the study of kinetic processes in molecular gases. It is also worth mentioning that experimental CRBS spectra of atomic gases have been interpreted on the basis of the simple BGK model mentioned above [10], properly modified to account for the optical lattice driving force [3]. In the following section we outline the method used for the simulations and point out the features of the GRID infrastructure that are exploited in order to obtain very precise results.

METHOD OF CALCULATION

Thermodynamic fluctuations in gases, provided the density is low enough that only bimolecular collisions are effective, are described by the Boltzmann equation. In order to get (estimates of) solutions of this equation the Direct Simulation Monte Carlo method is used in this work. Collisions between particles are modeled with the VSS model [16]. The model parameters can be adjusted in order for the simulated gas to reproduce accepted values for the transport coefficients and their temperature dependence. A homogeneous, equilibrium gas is simulated in a one-dimensional spatial domain. At the boundaries the system is in contact with gas in equilibrium [17]. As usual in DSMC simulations, the spatial cell width and the timestep are smaller than the corresponding characteristic molecular quantities. The density fluctuations are sampled at discrete space-time points, then Fourier transformed and squared to get the discrete power spectrum. It is easy to show that this simulated one-dimensional spectrum is simply proportional to that actually measured in the experiments. Also, since the density fluctuations are proportional to the gas density [7], the simulated fluctuations are equal to the real fluctuations to within the constant weight of the simulated particles. Therefore, the spectrum sampled by the simulation is exactly equivalent, to within normalization factors, to the spectrum measured in light scattering experiments.
In order to reduce the statistical scatter inherent in the particle simulation method ensemble averaging of the results is performed by averaging the results of many independent runs. This procedure also allows to estimate the variance of the results with respect to the statistical scatter. This procedure is amenable to implementation on a computational GRID [15]. In the worst case of calculations in the hydrodynamic limit, a single run takes about 10 hr of CPU time. The GRID infrastructure allows hundreds of runs to be performed simultaneously thus reducing drastically the global computational time. Provided the typical requirements of DSMC simulations are met, results are not very sensitive to the particular values chosen for the simulation parameters. The actual values used for this study are reported in Table 1.

Figure 1 shows the level of accuracy obtained in the simulation of the fluctuation spectrum in the hydrodynamic

TABLE 1. Recommended simulation parameters.

Parameter	Value
Mean free path to cell width ratio	5
Mean free time to timestep ratio	20
Average number of simulated particles per cell	10
Boundary conditions	Reservoir
Output to minimum wavelength ratio	32
Number of sampled spatial points	4096
$\omega_{\max}/c_0 k_{\max}$	1.5
Number of sampled time points	4096

regime (□=5.12 □m, y=9.0) as compared to the theoretical result [9]. In this case the calculation is particularly demanding since the simulated volume must extend for many mean free path lengths in order to sample fluctuations of the required wavelength (cfr. Tab. 1). A moderate number of particles can be used for the simulation since the spectrum is not sensible to this parameter, but results from many independent runs have to be averaged in order to get the required accuracy. In the present example 64 runs where necessary to obtain the results reported in fig. 1 which amount to approximately 640 hrs of CPU time. In the following section, the predictions of different theoretical models for the light scattering spectra of Argon in the kinetic regime are compared and discussed.

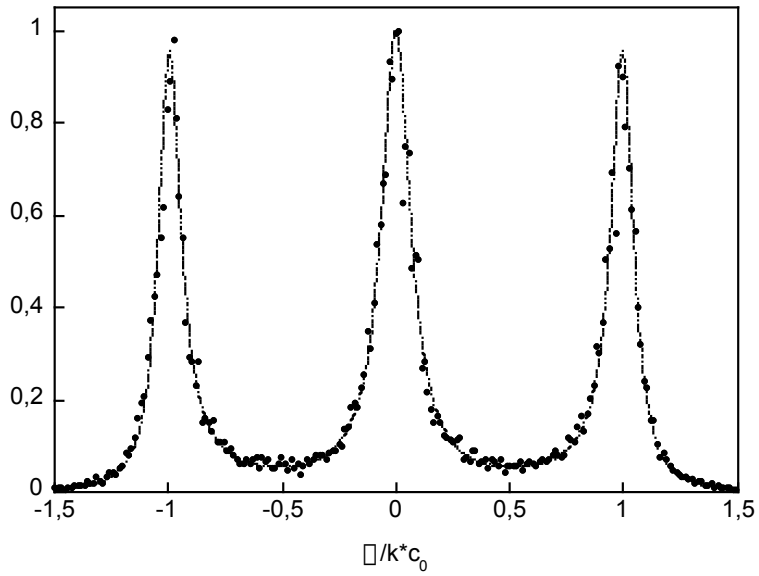

FIGURE 1. Light scattering spectrum of Argon as obtained by DSMC (symbols) and the continuum hydrodynamic theory [9]. Argon gas at 1atm and 292K. The probed wavelength is □=5.12 □m and the collisional parameter (cfr. eq. (4)) y=9.0.

RESULTS

Calculations have been performed for a monoatomic gas in equilibrium. The gas is Argon at p=101325 Pa and T=292 K. Other parameters used in the simulation are particle masses and the parameters of the VSS model. The latter are taken from the data reported in [14].

In this study we focus on the simulation of the fluctuation spectra in the kinetic regime where a general solution is still lacking. The collisional regime is specified by the ratio between the probed wavelength and the mean free path. Different definitions are possible. We adhere to the convention used in [3]:

$$y = \frac{8}{3\sqrt{2\pi}} \frac{\rho_0 \sqrt{k_B T_0 / m}}{\mu k} \tag{4}$$

where ρ_0 is the equilibrium density, μ is the shear viscosity and k is the fluctuation wavenumber.

The collisionless limit corresponds to $y \to 0$, whereas the hydrodynamic limit, $k \to 0$ is approached as $y \geq 5$. In the collisionless regime, where the fluctuation wavelength is much smaller that the mean free path, the spectrum of the spontaneous fluctuations is a Gaussian distribution whose width is determined by the gas temperature [7].

At much larger wavelengths, the system behaves like a continuum described by the hydrodynamic theory [8, 9]. In this regime, the spectrum is composed of a central peak, whose width is determined by the thermal conductivity coefficient, and two symmetric peaks, whose position is determined by the speed of sound and whose width is determined by the sound absorption coefficient given by (for monoatomic gases):

$$\Gamma = \frac{1}{2}\left[\frac{4}{3}\frac{\mu}{\rho_0} + (\gamma - 1)\frac{\kappa}{\rho_0 c_p}\right] k^2 \tag{5}$$

where γ is the specific heat ratio and $\alpha = \frac{\lambda}{\rho_0 c_p}$ the thermal diffusivity.

The shape of the spectrum is therefore determined by a combination of thermal conductivity and viscosity coefficients.

In the intermediate, or kinetic, regime the spectra are expected to shift continuously from the collisionless Gaussian form to the hydrodynamic limit. They are therefore expected to depend on the same physical properties of the gas, namely thermal conductivity and viscosity. The importance of this remark will become apparent later when we discuss the results of the BGK kinetic equation for this problem.

In the kinetic regime, the governing equations are not amenable to a analytical solution. However, by modeling the collision integral with a simple relaxation term it is possible to obtain a tractable model. The model developed in [10] and then extended to the CRBS case in [3] makes use of a BGK model equation linearised around the equilibrium solution. The model depends on a single adjustable parameter, which is a characteristic relaxation time of the instantaneous distribution function towards the local equilibrium distribution. By applying a Chapman-Enskog expansion to the BGK model equation it is possible to obtain the hydrodynamic limit and therefore to express the relaxation parameter in terms of the gas transport coefficients [10]:

$$\lambda = \frac{5}{4} k \frac{2 n k_B T}{m \omega} \tag{6}$$

$$\mu = \frac{n k_B T}{\omega} \tag{7}$$

From these relations it is evident that, within this model, viscosity and thermal conductivity cannot be matched independently. While this linearised model is amenable to analytical solution, the full BGK kinetic equation is not. In order to compare the solutions of the linearised model with the solutions of the full BGK model, we use a particle simulation method for the latter [5].

Simulations are carried out for Δ=5 10^{-5} cm or y=0.878.

Figure 2 shows the comparison of the DSMC results with the results of this simplified model. Several curves of the model for values of the model parameter of 0.05, 0.07, 0.09, 0.1, 0.12, 0.15 are reported. As the model parameter increases the spectrum changes from the Gaussian behaviour of the collisionless limit to the behaviour characterized by well defined sound propagation peaks. The values that reproduce the gas viscosity and thermal conductivity are 0.1 and 0.15, respectively. It is apparent how the model cannot reproduce the DSMC results by any possible value of the model parameter.

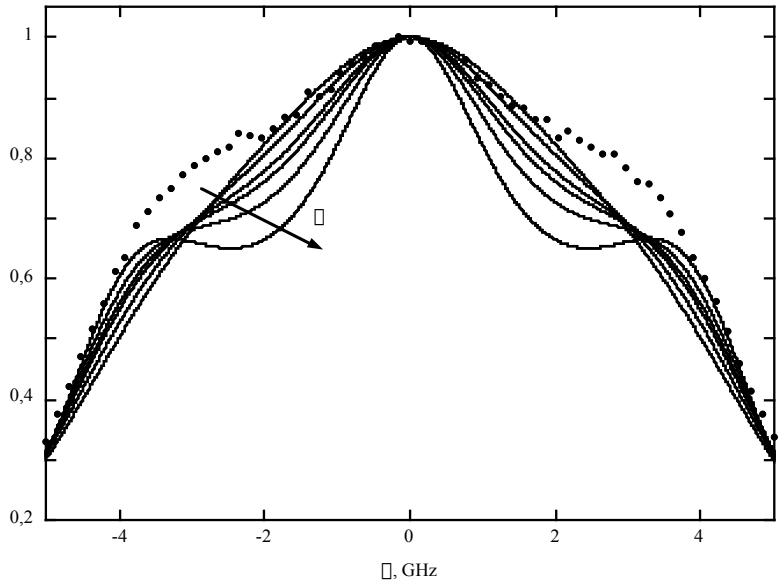

FIGURE 2. Light scattering spectrum of Argon as obtained by DSMC (symbols) and a linearised BGK model [10]. The curves reported have been obtained for the following values of the model parameter: Δ=0.05, 0.07, 0.09, 0.1, 0.12, 0.15.

It could be argued that the linearised model is very crude for application in the kinetic regime and that a direct solution of the full BGK eqution could give more correct results.
In this case the equation to be solved is:

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\cdot\frac{\partial}{\partial \mathbf{r}}\right) f = \nu \cdot \left(f_{eq} - f\right) \tag{8}$$

where f_{eq} is the equilibrium distribution at the instantaneous density and temperature.

Results obtained by this model are compared in fig. 3 with the analogous results obtained by the simplified model with the same model parameters.

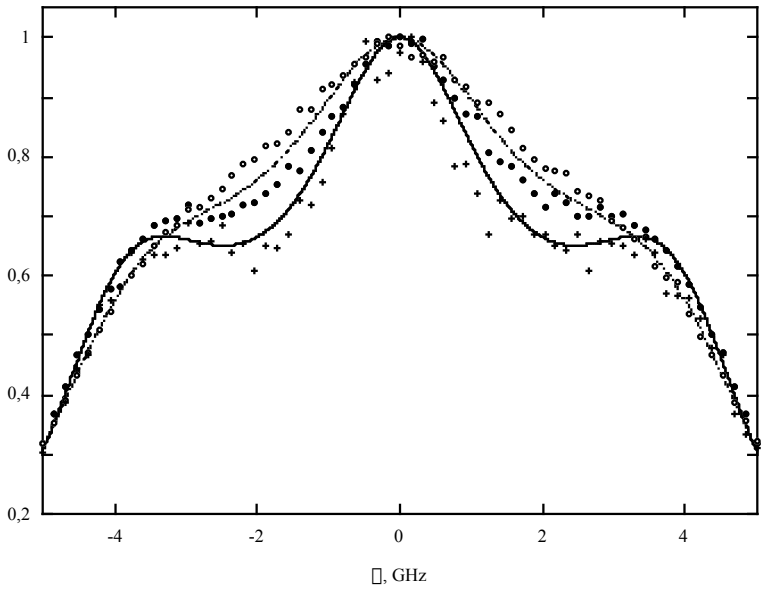

FIGURE 3. Light scattering spectrum of Argon as obtained by a linearised BGK model [10] (lines) and a particle solution of the full BGk equation (symbols) [5]. The curves reported have been obtained for the following values of the model parameter: =0.1, 0.15, 0.17 (symbols only).

It is apparent that, for fixed relaxation parameter, the simplified model forces a faster relaxation towards equilibrium so that the spectrum exhibits a more pronounced hydrodynamic behaviour. For example, the behaviour of the simplified model for =0.15, is obtained by the full model for =0.17. Nonetheless, the spectra obtained by the two models are completely equivalent and they describe the same physics.

Finally, in fig. 4 we compare DSMC results with full BGK results and try to adjust the free parameter of the BGK model in order to fit the DSMC results. Although better than the simplified model, BGK results cannot reproduce the features of the DSMC spectrum.

As mentioned before, the spectrum is determined by a combination of the thermal conductivity and viscosity properties of the gas system. But the BGK kinetic equation fails to reproduce correctly both physical properties and is therefore not able to simulate all the details of the spectrum.

CONCLUSIONS

Particle models exploiting the computational capabilities of GRID infrastructure are able to simulate light scattering spectra of gaseous systems in a wide range of collisional regimes with unprecedented accuracy. This allows the resolution of differences in the predictions of different theoretical models that are not amenable to analytical treatment. In particular, limitations of the BGK kinetic model with respect to the DSMC numerical experiments in the estimation of the light scattering spectra of atomic Argon in the kinetic regime have been clearly identified.

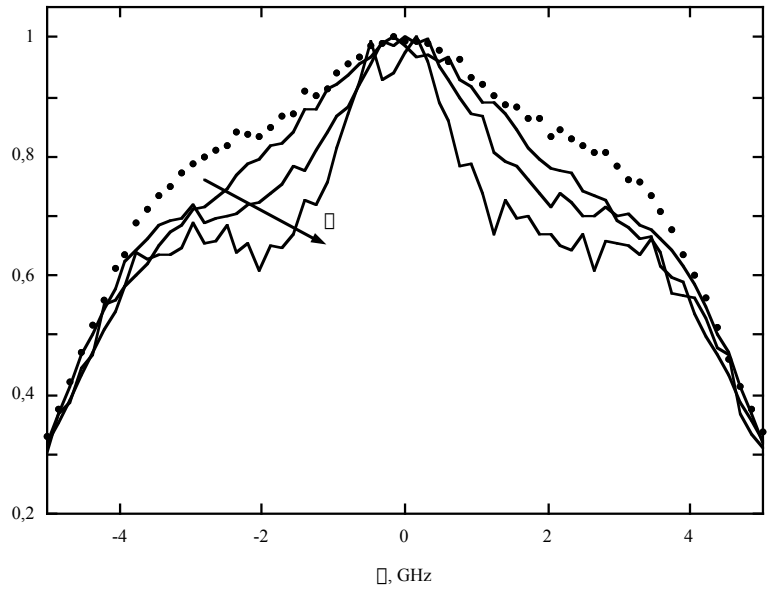

FIGURE 4. Light scattering spectrum of Argon as obtained by DSMC (symbols) and the full BGK model [5] (lines). The curves reported have been obtained for the following values of the model parameter: =0.1, 0.15, 0.17.

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