

INTERCOLLISIONAL INTERFERENCE EFFECTS IN COLLISION-INDUCED LIGHT SCATTERING

J. Courtenay Lewis and J. Van Kranendonk

Department of Physics, University of Toronto, Toronto, Ontario, Canada

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A general kinetic theory of intercollisional interference effects has been developed. Applied to collision-induced light scattering in rare gases, it predicts a peak at the incident frequency, with a height of about 25% of the intracollisional peak and a width proportional to the density.

Radiation processes occurring during successive binary collisions in pressure-induced infrared absorption¹ and in collision-induced light scattering² are not independent but show strong interference effects which originate in the correlations existing between successive intermolecular force pulses experienced by a given molecule in a gas.³ In induced absorption, these interference effects are negative and lead to a dip in the profiles of the Q branches of the translational and vibrational bands.⁴ This dip is located at the band origin, extends down to practically the zero absorption line, and has a width proportional to the density.¹ It arises from the fact that the dipole moments induced in two successive binary collisions are not randomly oriented but are most likely pointing in opposite directions. In collision-induced scattering, on the other hand, the interference is expected to be a constructive one and to lead to a peak at the center of the intramolecular spectrum of the scattered light. The induced polarizability responsible for the scattering² is even under inversion, and the negative correlation existing between the induced dipole moments in induced absorption therefore implies a corresponding positive correlation between the induced polarizability tensors. An analytic treatment of this effect based on kinetic theory is outlined here and an estimate of the magnitude of the effect is given.

Let $I_{\parallel}(\omega)$ be the intensity, at frequency ω from the incident frequency, of the light scattered in the x direction and polarized in the z direction, from a beam incident along the y direction and polarized in the x direction. For densities such that the mean free path is small compared with the wavelength and low enough that binary collisions predominate, the correlation-function expression for the scattered intensity is

$$I_{\parallel}(\omega) = \kappa \int_{-\infty}^{\infty} e^{i\omega\tau} \left\langle \sum_{i,j} \sum_{l,m} B(\vec{\mathbf{R}}_{ij}(t)) \times B(\vec{\mathbf{R}}_{lm}(t+\tau)) \right\rangle_t d\tau. \quad (1)$$

$\vec{\mathbf{R}}_{ij}(t)$ is the intermolecular separation of the

pair i, j at time t , $\kappa = \omega_0^4 E_0^2 / 2c^4 R_0^2$, and

$$B(\vec{\mathbf{R}}) = \alpha_x \alpha_z \beta(R); \quad (2)$$

α_x and α_z are direction cosines of $\vec{\mathbf{R}}$, $\beta(R)$ is the anisotropy of the polarizability of the pair, and R_0 is the distance from the scatterer.

The spectrum (1) consists of a broad intracollisional component with a width $\approx \tau_d^{-1}$, where τ_d is the duration of a collision, upon which is superposed the intercollisional component whose width is roughly τ_c^{-1} , where τ_c is the time between collisions. At not too high densities ($\tau_c \gg \tau_d$), the intracollisional spectrum is flat in the region where the intercollisional contribution is appreciable. To calculate the latter, we may therefore put

$$B(\vec{\mathbf{R}}(t)) = \bar{B}(\vec{\mathbf{k}}, g, b) \delta(t - t_0) \quad (3)$$

with

$$\bar{B}(\vec{\mathbf{k}}, g, b) \cong k_x k_z \bar{\beta}(g, b),$$

where $\bar{B}(\vec{\mathbf{k}}, g, b)$ is the time integral of $B(\vec{\mathbf{R}}(t))$ for a collision of type⁵ $\vec{\mathbf{k}}, g, b$, $\bar{\beta}$ is the time integral of $\beta(R(t))$, and t_0 is the time of closest approach. To obtain the intracollisional spectrum at higher frequencies, one must replace Eq. (3) by a more realistic expression. This problem has been discussed by Levine and Birnbaum², and by Sears.⁶

For a gas containing N molecules, we obtain from (1) and (3)

$$I_{\parallel}(\omega) = 4\kappa N \nu_c [I_{\text{intra}}(\omega) + I_{\text{inter}}(\omega)], \quad (4)$$

where

$$I_{\text{intra}}(0) = \frac{1}{2} \langle \bar{B}_i \bar{B}_i \rangle_c, \quad (5)$$

$$I_{\text{inter}}(\omega) = 2\text{Re} \sum_{n=1}^{\infty} \left(\frac{\nu_c}{\nu_c + i\omega} \right)^n \langle \bar{B}_i \bar{B}_{i+n} \rangle_c. \quad (6)$$

The subscript $j = i$, $i+n$ refers to the j th collision experienced by a given molecule from some reference time, ν_c is the collision frequency, and $\langle \dots \rangle_c$ denotes an average over all possible sequences of $n+1$ collisions $i, \dots, i+n$. The simple form of the frequency dependence in Eq. (6) arises from the assumption that

the collisions of a given molecule have a uniform random distribution in time. This is equivalent to assuming molecular chaos and velocity independence of the collision frequency. Taking the velocity dependence of the latter into account does not affect $I_{\text{inter}}(0)$ and alters the frequency dependence only insignificantly.

We denote the velocities of a molecule before and after collision by \vec{c}_1 and \vec{c}_1' , and set $\vec{C} = \vec{c}_1' - \vec{c}_1$. ξ is the angle between \vec{c}_1 and \vec{c}_1' , η is the angle between \vec{c}_1 and \vec{C} , and η' is the angle between \vec{c}_1' and \vec{C} . If we assume that $\bar{\beta}$ depends only on c_1 , c_1' , and ξ , a recursion relation can be established connecting $\langle \bar{B}_i \bar{B}_{i+n} \rangle_c$ and $\langle \bar{B}_i \bar{B}_{i+n+1} \rangle_c$ which enables one to sum Eq. (6), yielding

$$I_{\text{inter}}(\omega) = \frac{2}{15} \text{Re} \left[\frac{\nu_c}{\nu_c + i\omega} \int_0^\infty dc_1 P(c_1) \int_0^\infty dc_1' \int_0^\pi P(\xi, c_1' | c_1) P_2(\cos \eta') G(\omega, c_1') \bar{\beta} \sin \xi d\xi \right], \quad (7)$$

where $G(\omega, c_1)$ is the solution of the equation

$$G(\omega, c_1) = A(c_1) + \frac{\nu_c}{\nu_c + i\omega} \int_0^\infty dc_1' \Delta(c_1' | c_1) G(\omega, c_1'), \quad (8)$$

$$A(c_1) = \int_0^\infty dc_1' \int_0^\pi P(\xi, c_1' | c_1) P_2(\cos \eta) \bar{\beta} \sin \xi d\xi, \quad (9)$$

and

$$\Delta(c_1' | c_1) = \int_0^\pi P(\xi, c_1' | c_1) P_2(\cos \xi) \sin \xi d\xi. \quad (10)$$

The height of $I_{\text{intra}}(\omega)$ at zero frequency is

$$I_{\text{intra}}(0) = (1/30) \int_0^\infty dc_1 P(c_1) \int_0^\pi dc_1' \int_0^\pi P(\xi, c_1' | c_1) \bar{\beta}^2 \sin \xi d\xi. \quad (11)$$

$P(c_1)$ is the distribution of c_1 , $P(\xi, c_1' | c_1)$ is the conditional distribution of ξ and c_1' given c_1 , and P_2 is a Legendre polynomial. Similar equations describe the intercollisional interference effect in induced absorption, but with P_1 in place of P_2 . Because of the more rapid variation of P_2 , the interference effect in light scattering is more sensitive to the details of the intermolecular interaction than in induced absorption.

The integral equation (8) incorporates the correlations between an arbitrary number of successive collisions, i.e., all terms in Eq. (6) are included, and constitutes the main result of our theory. In conjunction with Eq. (7), this result makes possible a detailed calculation of the intercollisional component in induced absorption and light-scattering spectra. The most important parameters characterizing this component are the ratio $\mathcal{R} \equiv I_{\text{inter}}(0)/I_{\text{intra}}(0)$, and the width $\Delta\omega_{\text{inter}}$ of $I_{\text{inter}}(\omega)$. For induced absorption, \mathcal{R} is very nearly equal to -1 as a result of the approximate proportionality of the induced dipole moment and the intermolecular force.⁴ To estimate the value of \mathcal{R} for collision-induced light scattering, we assume that $\beta(R)$ is proportional to the magnitude of the intermolecular force, or $\bar{\beta} = \text{const} \times C$, and we use the hard-sphere expression for $P(\xi, c_1' | c_1)$. Then Eqs. (7)-(11) can be evaluated, giving $\mathcal{R} \approx +0.25$. The interference in this case is, therefore, predicted to be constructive and to make an appreciable contribu-

tion to the total peak height.

The correlation between force pulses on a molecule in a gas is strongest for immediately successive collisions. One would expect, therefore, that the first ($n=1$) term of Eq. (6) would be the largest. In our calculation it is in fact the dominant term, contributing 65% of $I_{\text{inter}}(0)$. Hence $I_{\text{inter}}(\omega)$ should be predominantly Lorentzian but with appreciable deviations due to the terms with $n > 1$ in Eq. (6). Then $\Delta\omega_{\text{inter}} \approx 2\nu_c$. As ν_c is proportional to the density (at not too high densities), the width $\Delta\omega_{\text{inter}}$ should be approximately proportional to the density, in agreement with the experimental results for induced absorption.¹

As discussed in previous work,⁴ the great advantage of studying the intercollisional component in induced absorption and light-scattering spectra is that the entire Fourier spectrum of the underlying correlation function is open to observation, whereas in the case of other transport properties, such as diffusion and heat conduction, only the $\omega=0$ component can be measured. Detailed measurements of the density and temperature dependence of $I_{\text{inter}}(\omega)$ in induced infrared spectra are under way.⁷ In the case of light scattering, the observation of $I_{\text{inter}}(\omega)$ is more difficult because of the presence of the strong incident intensity at $\omega=0$, and it will probably be necessary to go to considerably

higher densities, of the order of 500 amagat, than used so far.² We point out that the analysis of the light-scattering spectra is complicated by the fact that at small ω an appreciable contribution to the scattering may be due to the long-range dipole-dipole interaction⁸ rather than to the short-range intermolecular interaction.² As a first step, it would be very interesting if the predicted constructive nature of the interference could be verified experimentally.

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EFFICIENT MODULATION COUPLING BETWEEN ELECTRON AND ION RESONANCES IN MAGNETOACTIVE PLASMAS*

Alfred Y. Wong, Dan R. Baker, and Newell Booth

Department of Physics, University of California, Los Angeles, California 90024

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Modulation coupling between electron upper-hybrid modes and ion modes propagating nearly perpendicular to the axial magnetic field is shown to be effective in collisionless and semicollisionless regimes. A complete probeless feedback stabilization scheme based on such mode coupling is described.

We wish to report a new and efficient "modulation coupling" scheme between electron and ion resonances over a wide density range ($10^8 \lesssim n \lesssim 10^{11} \text{ cm}^{-3}$) and wide ion-frequency regime $1 \text{ kHz} \lesssim f_i \lesssim 200 \text{ kHz}$ in a magnetically confined plasma. This coupling scheme, which does not appear to depend significantly on collisional processes, permits one to excite or suppress ion resonances in plasmas "remotely" without either grids or probes while retaining the significant property of spatial selectivity. We also demonstrate that by means of such a coupling scheme, low-frequency ion instabilities in hot or rarefied plasmas can be controlled efficiently by microwave beams of relatively low power. This has important implications in controlled fusion and excitation of waves in space plasmas by ground stations. The modulation coupling in the stable regime could be relevant to modulation instabilities which are of increasing theoretical interest.

The experiments were performed in a highly ionized potassium plasma produced in a Q device ($T_e \approx T_i \approx 0.2 \text{ eV}$) operated in a symmetrical manner as in Fig. 1(a). In the direction perpendicular to \vec{B} , the electron electrostatic modes are the Bernstein modes at the upper hybrid frequency $\omega_{UH}^2(r) = \omega_p^2(r) + \omega_{ce}^2$, while the ion modes are the electrostatic ion cyclotron waves and drift

waves whose damping rates can be easily controlled by varying the magnetic field or the plasma density. The resonant character of upper-hybrid modes¹ and the ion cyclotron modes² arises from a combination of cyclotron motion and collective effects, while drift modes³ derive their resonant behavior from the density gradient and the periodicity in the azimuthal direction. Low microwave power of the order of a few milliwatts and frequency 2-4 GHz is radiated upon the plasma column (5 cm in diameter and 80 cm in length) by waveguides located outside the chamber. The microwave is in the extraordinary mode along a direction perpendicular to the magnetic field. The microwave frequency is chosen to correspond to the upper hybrid frequency in the range of the magnetic field (1-1.5 kG) and density at which ion resonances have small damping rates. It has been demonstrated experimentally that an evanescent layer normally shields the upper hybrid resonance in the radial direction.¹ This can be overcome by either imposing a gradient in the external magnetic field or making the microwave wavelength exceed the plasma radius. We have chosen the latter because a uniform B field is essential in our identification of the resonant modes. The electron resonances were identified by a forward-scattering technique⁴ and by moni-