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Analysis of the $R_1(J)$- and $P_1(J)$-Branch Absorption Spectrum of HD–Rare-Gas Mixtures: An Example of Positive Intercollisional Interference

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A theory for the intensities and shapes of fundamental band $R_1(J)$ and $P_1(J)$ lines of HD under pressures of rare gases is presented, with specific application to the $R_1(1)$ line of HD-Kr. The absorption contours principally consist of a broad (~100 cm^{-1}) feature representing the ordinary intracollisional dipole intensity, and narrow components arising from the small HD permanent dipole moment function; permanent-dipole–collisionally-induced-dipole interference; and positive intercollisional interference between dipoles induced in successive collisions.

For many years now, pressure-induced vibration-rotation absorption spectra of molecular hydrogen and its isotopes have been of considerable interest both experimentally and theoretically. One of the outstanding features of these spectra as induced by foreign gases is the existence of a pronounced dip[1] in the intensity contour at the position of the Q branch in the fundamental band $(v=0,\ J)\rightarrow(v=1,\ J)$. This dip has been interpreted[2] with success in terms of a destructive interference effect associated with the relative orientations of the transition dipoles induced during different (primarily successive) collisions. In addition to these destructive intercollisional effects Lewis and Van Kranendonk[3] have predicted positive intercollisional interference effects (represented by the appearance of peaks rather than dips) in the depolarized components of collision-induced Rayleigh scattering by gases. Such effects have not yet been observed, however.

Superposed on the broad features characteristic of the H_2-induced dipole spectrum, sharp peaks have been observed at the positions of the pure rotation lines[4] $(0,J)\rightarrow(0,J+1)$, the fundamental band[5,6] $R_1(J)$ branch $(0,J)\rightarrow(1,J+1)$ and $P_1(J)$ branch $(0,J)\rightarrow(1,J-1)$ lines, and in at least three overtone bands[5] $(v=0\rightarrow2,3,4)$ in pure HD at various pressures. These lines have been attributed,[4-7] at least partially, to the existence of the very small permanent dipole which gives rise to weak transitions. Even more recently, a similarly sharp feature has been observed[8,9] at the $R_1(1)$ line position in HD-Kr and HD-Xe mixtures which is too intense to be explained solely in terms of the permanent dipole moment function. Poll, Tipping, Prasad, and Reddy[9] (henceforth referred to as I) have ascribed its intensity to an interference between the permanent and collisionally induced dipole moments. However, they did not calculate the frequency distribution of this contribution, which is necessary for comparison with spectroscopic observations, nor have they obtained all of the contributions to the sharp feature.

The purpose of this paper is to identify in detail the mechanisms responsible for producing $R_1(J)$ and $P_1(J)$ features in HD-Kr (and, in principle, HD mixtures with lighter rare gases) and to calculate their shape and intensity. According to our findings, these lines each consist of five distinct contributions, as listed later in this paper. In particular, there are three distinct contributions to the sharp feature yielding identical line shapes in the impact limit, including an intercollisional interference contribution, proportional to $\rho_{HD}\rho_{Kr}^2$, as well as the collisional-dipole–permanent-dipole interference term mentioned above, which is proportional to $\rho_{HD}\rho_{Kr}$. While the experiments of Prasad and Reddy indicate only that an enhancement in line strength due to collisions exists, our numerical calculations reveal that the intercollisional term is competitive with the permanent-dipole–collisional-dipole interference term over the pressure range studied. Further experimental work is necessary to isolate the effects of these contributions.

Interaction energies and dipole moments in HD-Kr *collisions.*—The interaction between HD and a rare-gas atom is dominated by the spherically symmetric van der Waals interaction potential, an interaction proportional to the displacement $\vec{x}$ $(=\vec{r}/6)$ of the center of mass from the electrical centroid in HD,[10] and a term associated with the "elongation" of the hydrogen molecule,[11]

$$U(\vec{r},\vec{R}) = U_0(r,R) + x[\partial U_0(r,R)/\partial R]P_1(\cos\gamma) + U_2(r,R)P_2(\cos\gamma), \tag{1}$$

and the HD center of mass→rare-gas displacement $\vec{R} = (R,\Theta,\Phi)$ (see Fig. 2 of I).

Associated with each term in Eq. (1) there exist contributions to the induced dipole (νth spherical component),

$$\mu_{I_\nu}(\vec{r},\vec{R}) = \left(\frac{4\pi}{3}\right)^{1/2}\left(A_{10}(r,R) + x\,\frac{\partial A_{10}(r,R)}{\partial R}P_1(\cos\gamma)\right)Y_{1\nu}(\Theta,\Phi) + [\sim P_2(\cos\gamma)] \tag{2}$$

in the notation of I to an approximation consistent with Eq. (1). The $A_{10}(r,R)$ term is independent of the orientation of HD, while the "elongation" term varies as $P_2(\cos\gamma)$. Only the $P_1(\cos\gamma)$ term, which again arises from the displacement of centers in HD, is important in the present problem. Together with the weak permanent dipole moment function, it gives rise to the $\Delta J = \pm 1$ transitions of present interest. Expressing $\cos\gamma$ in terms of θ, φ, Θ, and Φ, this term becomes

$$\mu_{I_\nu} = \frac{1}{3}x\,\frac{\partial A_{10}(r,R)}{\partial R}\times\begin{cases}(4\pi/3)^{1/2}Y_{1\nu}(\theta,\varphi), & \text{(3a)}\\ -4(2/3)^{1/2}\sum_{M=-2}^{2}C(121;(\nu-M)M)Y_{1(\nu-M)}(\theta,\varphi)Y_{2M}(\Theta,\Phi), & \text{(3b)}\end{cases}$$

which should be compared with the form of Eqs. (1), (5) of I. To expression (3a) should be added the νth component of the small-permanent-dipole operator, $(4\pi/3)^{1/2}P_A(r)Y_{1\nu}(\theta,\varphi)$ [Eq. (2) of I]. This term and (3a) together lead to the sharp lines.[12] In addition, (3a) above leads to broad components which underlie the sharp features. The term (3b) has as its principal effect the augmentation of these broad features, having twice the effect of (3a) in this regard.[13] Setting aside (3b) we now write the total operator in the form

$$\mu_\nu = [P_I(r,R) + P_A(r)](4\pi/3)^{1/2}Y_{1\nu}(\theta,\varphi), \tag{4}$$

with

$$P_I(r,R) = \tfrac{1}{3}x\partial A_{10}(r,R)/\partial R. \tag{5}$$

Intensity distribution resulting from $\mu_\nu(\vec{r},\vec{R})$.—The absorption coefficient as a function of angular frequency is expressed in terms of the dipole-dipole correlation function,[9]

$$\alpha(\omega) = \omega\kappa_J\int_{-\infty}^{\infty}d\tau\, e^{-i\omega\tau}\{\langle 0,J|P_I(t)+P_A|1,J+1\rangle_t\langle 1,J+1|P_I(t+\tau)+P_A|0,J\rangle_{t+\tau}\}_{\text{ens}} \tag{6}$$

with $\kappa_J = \frac{4}{3}\pi^2 n_{\text{HD}}a_0^2\alpha_F P_J C(J\,1(J+1);00)^2$. The subscripts t and $t+\tau$ denote that the states $(0,J)$, etc., entering the radial matrix elements are[14] Heisenberg states at t, and collisionally smeared states at $t+\tau$ having developed from the Heisenberg states during the interval $t\to t+\tau$. The symbol $\{\,\}_{\text{ens}}$ indicates the ensemble average over classical collisional paths.

The intercollisional effect of importance in producing sharp lines arises from the fact that while individual collisions are isolated in time and are statistically independent, the phase of the transition dipole is highly correlated throughout a collision sequence,[12] persisting for times limited only by collisional broadening processes.[15] Pressure broadening exhibits the effects of small phase shifts suffered in each of a large multitude of binary collisions, while the phase shifts associated with collisions in progress at t and $t+\tau$ may be neglected without significant error.[16] Accordingly, pressure-broadening influences are quite independent of collisional dipole production, so that one may take separate ensemble averages in describing these influences. Abbreviating the radial matrix element amplitudes by $p_I{}^0(t)$ and $p_A{}^0$, Eq. (6) thereby reduces to the form[17]

$$\alpha(\omega) = \omega\kappa_J 2\,\text{Re}\int_0^{\infty}d\tau\,\exp[i(\omega_0-\omega)-\Gamma/2]\tau\{p_I{}^0(t)+p_A{}^{0*}p_I{}^0(t+\tau)+p_A{}^0)\}_{\text{ens}} \tag{7}$$

in the impact limit with ω_0, $\text{Re}(\Gamma/2)$ and $\text{Im}(-\Gamma/2)$ being the unperturbed transition frequency, and the half width at half intensity of the resulting Lorentzian contour and the pressure shift.

The ensemble average $\{[P_I^0(t)+p_A^0]^*[p_I^0(t+\tau)+p_A^0]\}_{\rm ens}$ can be separated as follows. (a) The largest contribution in total intensity, for all but the very lowest pressures, arises from the intracollisional term—that is, from the self-correlation of the dipole induced in each individual collision—and leads to a frequency spectrum centered at ω_0 having width of order $2\tau_d^{-1}$. Its intensity depends on the frequency of binary encounters and is given by Eq. (4b) of I, although as explained in the discussion following Eq. (3), this result ultimately must be multiplied by a factor of 3. (b) A mix consisting of (1) the permanent-dipole contribution, (2) the collisional-dipole–permanent-dipole interference, and (3) the intercollisional interference in which $p_I^0(t)$ and $p_I^0(t+\tau)$ are induced in different collisions has τ-independent magnitude and therefore gives rise to the pressure-broadened line shape. (c) Another intercollisional interference contribution results from the fact that the collisional speed at time $(t+\tau)$ is not independent of that characterizing the collision in progress at time t. In practice the HD molecular speed significantly changes its magnitude only after a few momentum-exchanging collisions with heavier rare-gas atoms. Accordingly for HD-Kr this (presumably weak) component is considerably broader than the pressure-broadened line shape while still being narrow compared with intracollisionally generated features.

The various components, their widths, central frequencies, and integrated absorption coefficients, $\int\alpha(\omega)\omega^{-1}d\omega$, together with density dependences are listed below for the $R(J)$ transitions. Here, all effects arising from HD-HD collisions are ignored.

(1) The intracollisionally induced dipole intensity having large density-independent linewidth ($\sim 2\tau_d^{-1}$), centered approximately at the unperturbed line position having strength $12\pi n_{\rm Kr}a_0^3\kappa_J \times\int_0^\infty[p_I^0(R)]^2 g(R)R^2\,dR$ and density dependence $\sim\rho_{\rm HD}\rho_{\rm Kr}$. [This includes the contribution arising from the $L=2$ terms indicated by Eq. (3b).]

(2) A contribution to the sharp feature which is characterized by the collisionally broadened line shape for an allowed transition {Lorentz contour centered at $[\omega_0-{\rm Im}(\Gamma/2)]$ having linewidth ${\rm Re}(\Gamma)$, Γ being proportional to $\rho_{\rm Kr}$} arising from the small HD permanent dipole, having strength $\kappa_J(p_A^0)^2$, proportional simply to $\rho_{\rm HD}$.

(3) The allowed collisionally induced interference contribution to the sharp line (having the same line shape and position) with strength $2\kappa_J p_A^0[4\pi n_{\rm Kr}a_0^3\int_0^\infty p_I^0(R)g(R)R^2\,dR]$, proportional to $\rho_{\rm HD}\rho_{\rm Kr}$.

(4) The intercollisional interference contribution to the sharp line (again having the same line shape and position) with strength $\kappa_J[4\pi n_{\rm Kr}a_0^3 \times\int_0^\infty p_I^0(R)g(R)R^2\,dR]^2$, proportional to $\rho_{\rm HD}\rho_{\rm Kr}$.

(5) The (presumably weak) intercollisional feature associated with the persistence of HD molecular speeds, having width ~ (several collision times)$^{-1}$, proportional to $\rho_{\rm Kr}$, and strength $\kappa_J(4\pi n_{\rm Kr}a_0^3)^2\int_0^\infty dc\,P(c)\{[\int_0^\infty g_c(R)p_I^0(R)R^2\,dR]^2-[\int_0^\infty g(R)p_I^0(R)R^2\,dR]^2\}$, proportional to $\rho_{\rm HD}\rho_{\rm Kr}$, with $P(c)$ and $g_c(R)$ being, respectively, the HD molecular speed distribution and the HD-Kr pair distribution function for molecules having speed c.

It is significant that the intercollisional interference terms, which are proportional to $\rho_{\rm HD}\rho_{\rm Kr}^2$, do not in any way interfere with the binary terms, which are proportional to $\rho_{\rm HD}\rho_{\rm Kr}$. These terms are, instead, truly ternary in their origin (even though we have regarded them as being due to a succession of statistically independent binary events) as is signaled by the fact that mathematically their intensity is related to the intercollisional contribution to the dipole-dipole correlation function at $\tau=0$, which is simply the probability for the coincidence of binary events[18] (i.e., the probability for ternary collisions). It is easily shown, in this regard, that the narrow intercollisional component accounts for nominally half the total ternary integrated absorption.

Numerically, the ratio of the intergral $[4\pi n_{\rm Kr}a_0^3 \times\int_0^\infty g(R)p_I^0(R)R^2\,dR]$ to p_A^0 is 2.1 : 1.0 at 100 amagats Kr density[19]—as is compatible with the numerical analysis of I. Consequently, one may not neglect p_A^0 although the $(p_A^0)^2$ terms are definitely rather small in the range of Kr densities for which $R_1(1)$ has been experimentally observed. A numerical expression for the intensity of the sharp $R_1(1)$ line is $(\rho_{\rm HD})^{-1}\int\alpha(\omega)\omega^{-1}d\omega=0.90 \times10^{-8}[(1+4.2)\times10^{-2}\rho_{\rm Kr}+4.4\times10^{-4}\rho_{\rm Kr}^2]$ cm^{-1} amagat^{-1}, where the densities are expressed in amagats.[20] For ~ 100 amagat Kr densities, therefore, the predicted integrated absorption coefficient of the sharp $R_1(1)$ line is now approximately 70% greater than the experimentally determined values.[9] The experimental data may therefore suggest a somewhat reduced value for the average induced moment—however, not by so much that the quadratic term in $\rho_{\rm Kr}$ would lose its importance.

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[12]This arises from the fact that (3a) is completely independent of the angular position of the collision partners, as specified by (Θ, Φ). The phase of the transition dipole is governed solely by the internal dynamics of the HD molecules which remains unperturbed through many collisions.

[13]These broad components are difficult to isolate in the fundamental band, although Trefler and co-workers (Ref. 4) have observed their effects in the pure rotational spectrum of HD.

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[15]The interferences between transition amplitudes induced in separate collisions at different times is equivalent to that utilized in Ramsey's double-resonance experiments [N. F. Ramsey, *Molecular Beams* (Oxford, Clarendon Press, London, 1956)], which serve to increase the precision with which atomic frequencies may be measured by lengthening the effective observation times to those between interactions rather than those during interactions with an applied resonant field. In the present manifestation, it is the collisions themselves which (randomly and multiply) turn on and off the interaction between the HD molecule and the applied optical field.

[16]Significant phase changes introduced specifically during dipole-producing collisions can indeed alter the outcome of the present treatment, as will be shown in a future publication. These will be ignored in the present paper; however, the present treatment may ultimately be regarded as accurate only for the most inert rare gas collision partners.

[17]Because $\alpha(\omega)$ is real, the interval $-\infty < \tau \leq 0$ always yields the complex conjugate of the interval $0 \leq \tau < \infty$.

[18]The situation regarding the destructive intercollisional interference in the Q branch appears to be analogous in this regard.

[19]Based on numerical data supplied by J. D. Poll, private communication.

[20]It is assumed that for heavier rare gases $p_A{}^0$ and $p_I{}^0$ have the same sign since in each case, the dipole moment derivatives are parallel to r. For the lighter rare gases, the $p_I{}^0$ contribution may change sign.

Radiation Reaction as a Mechanism for Increasing Energy-Momentum of a Particle Interacting with a Laser Field

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It is shown, via explicit solution of the Lorentz-Dirac equation to second order, that radiation reaction is responsible for increasing the asymptotic energy-momentum of a charged particle being swept over by an intense pulse of laser radiation. The amount of increase is dependent upon the total electromagnetic energy in the laser pulse per unit area of wave front.

Recently, a great deal of attention has been focused on the ability of free electrons to emit, under certain prescribed situations, coherent radiation. The analyses made by Madey,[1] and Madey, Schwettman, and Fairbank,[2] of the stimulated emission of bremsstrahlung by free electrons