

A New Optical Method for the Determination of Interatomic Potentials

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A new method is described to obtain information on the true difference potential of interatomic interaction from the quasistatic wings of spectral lines broadened by neutral atoms. Based on the one-particle-approximation of the quasistatic theory of line broadening, it does not rely on any particular model function. Furthermore, from measurements of the temperature dependence of the line wings it may yield additional information on the potentials for the initial or final state of the line separately. The method is applied to measurements of the red wing of the mercury absorption line $6^1S_0-6^3P_1$, λ 2537 Å, perturbed by argon.

1. Introduction

The profile of a line broadened by neutral atoms reflects the potentials $V''(r)$ and $V'(r)$ for the initial and final state of the line as functions of the internuclear distance r between active and perturbing atom. To first approximation only the difference potential $V(r) = V'(r) - V''(r)$ is of importance for the formation of the line profile. For its determination the following procedure, which is also often used in molecular beam scattering, has been adopted so far in the literature on line broadening: The true difference potential, which is very complicated, is approximated by some simple analytical model function, for example of the Lennard-Jones type. With such a model function, containing a few adjustable parameters, the line profile is calculated on the basis of some proper line broadening theory and the unknown parameters fitted to part of the experimental data. If the potential function thus obtained is not consistent with all experimental data, the number of parameters is increased until consistency is established. The result of this step-by-step-procedure is satisfactory only, if the number of parameters is small compared to the number of linearly independent observables. As an example, the two constants of the Lennard-Jones(12,6)-potential may be determined from halfwidth and shift as functions of the perturber number density, as measured at sufficiently low density and at constant temperature. A consistency check of the potential is then possible by additional measurements of the density dependence at intermediate and higher den-

sities, of the temperature dependence of the line core and of the spectral intensity distribution in the line wing. A great deal of investigations of this type has been reported by a number of authors, in particular HINDMARSH¹, BEHMENBURG² and VAUGHAN³. The result was, that the L.-J.(12,6)-potential is in many cases a good first approximation to the true difference potential.

In this paper a new method for the determination of potential functions is described, which does not rely on any particular model function. With this method, which is based on the one-particle approximation of the quasistatic theory, information on the true difference potential is obtained directly from the line wings. Furthermore, from measurements of the temperature dependence of the wings it may yield additional information on the potentials for the initial or final state of the line separately.

In the following sections first the theoretical foundations of the method are outlined. Thereafter the method is applied to measurements of the red wing of the mercury line $6^1S_0-6^3P_1$, λ 2537 Å, perturbed by argon.

2. Theory

2.1. The Basic Intensity Formula

In deriving the intensity formula for the line wings on the basis of the quasistatic theory⁴⁻⁶ we make the following assumptions:

1) The active atom is surrounded by perturbing atoms, which are at rest relative to the active atom; this is equivalent with assuming, that the effects of collisions are negligible.

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2) Only the effect of the nearest perturber is taken into account; This means, that the number density of the perturbers is assumed to be sufficiently small.

3) The oscillator strength of the line under investigation is independent of r in the energy range under consideration.

4) There exists at most one value r_e , where the true difference potential $V(r)$ passes through an extremum.

We call $\Delta\nu = \nu - \nu_0$ the frequency perturbation (ν_0 = unperturbed frequency, ν = absorbed or emitted frequency), which is related to the difference potential by:

$$\Delta\nu(r) = (1/h) V(r) \quad (h = \text{Planck's constant}). \quad (1)$$

Furthermore we denote by $W(r) dr$ the probability of finding the nearest perturber in the interval between r and $r + dr$, causing a frequency perturbation between $\Delta\nu$ and $\Delta\nu + d(\Delta\nu)$. The intensity $I(\Delta\nu) d(\Delta\nu)$ absorbed or emitted in the frequency range between $\Delta\nu$ and $\Delta\nu + d(\Delta\nu)$, resulting from assumption 1), 2) and 4), depends on whether or not an extremum exists in $V(r)$.

Case 1: An extremum exists in $V(r)$. Then $V(r)$ becomes zero at $r = \sigma$.

$$I(\Delta\nu) d(\Delta\nu) = \begin{cases} W(r_1) dr_1 + W(r_2) dr_2 & \text{if } r > \sigma, \\ W(r) dr & \text{if } r < \sigma, \end{cases} \quad (2.1)$$

$$\sigma < r_1 < r_e < r_2.$$

Case 2: An extremum does not exist in $V(r)$.

$$I(\Delta\nu) d(\Delta\nu) = W(r) dr. \quad (2.2)$$

2.2. The Integration of the Intensity Formula

From the measured intensity distribution and known probability distribution we may then, in principle, obtain part of the function $\Delta\nu(r)$ and therefrom the difference potential $V(r)$, by simply integrating Eqs. (2). In order to get a one-to-one relation between $\Delta\nu$ and r in case 1, it has to be required, that in the range $r > \sigma$ the term $W(r_1) dr_1$ is negligibly small compared to the term $W(r_2) dr_2$. The requirement is fulfilled only for systems with $\sigma'' \gg \sigma$, so that $\sigma < r_1 < r_e$ falls into the region of large positive energies of the ground-state potential $V''(r)$, where the Boltzmann factor becomes very small. We then obtain from (2)

$$\int_{\Delta\nu_0}^{\Delta\nu} I(\Delta\nu) d(\Delta\nu) = \int_{r_0}^r W(r) dr. \quad (3)$$

Having chosen (to some degree arbitrarily) a value for $\Delta\nu_0$ within the validity range of the quasistatic theory, the value of r_0 is then determined by the boundary condition

$$\int_{\Delta\nu_0}^{\infty} I(\Delta\nu) d(\Delta\nu) = \int_{r_0}^0 W(r) dr. \quad (4)$$

With these values for the lower limits of the integrals $\Delta\nu(r)$ may be obtained from (3) by numerical integration.

2.3. The Probability Distribution

For $W(r) dr$ we find from assumption (2) and elementary laws of the theory of probability

$$W(r) dr = 4\pi r^2 n(r) dr \exp\left\{-\int_0^r 4\pi r^2 n(r) dr\right\}, \quad (5)$$

$$\int_0^{\infty} W(r) dr = 1.$$

The number density $n(r)$ of the perturbers in the neighborhood of the active atom is given by

$$n(r) = n_0 \exp\{-V''(r)/kT\} \cdot I(r); \quad (6)$$

n_0 denotes the number density in the absence of interaction. This is modified in the presence of interaction by the Boltzmann factor $\exp\{-V''(r)/kT\}$ and the relative number $I(r)$ of unbound perturbers

$$I(r) = \frac{\int_0^{\infty} \frac{1}{2} \cdot \exp\{-p^2/2kT\} 4\pi p^2 dp}{\int_0^{\infty} \exp\{-p^2/2kT\} 4\pi p^2 dp} \quad (7)$$

(μ , p reduced mass and momentum of relative motion of active and perturbing atom, respectively).

It is obvious, that in general the intensity distribution in the statistical wings does not only depend on the difference potential, but also on the ground state- (or excited state-) potential. Information on the latter, especially on its repulsive part, may be obtained from additional measurements of the temperature dependence of the intensity distribution in the wings.

3. Experimental Details and Results

3.1. The Absorption Tube

The mercury line λ 2537 Å was formed in absorption. The absorption tube was of conventional type, made of a quartz tube 30 cm long and 3 cm in diameter, with plane quartz windows at the end. Spectrally

pure argon at a number density of $1.26 \times 10^{19} \text{ cm}^{-3}$, together with a droplet of mercury were introduced into the highly evacuated tube, which was melt off thereafter. The maximum temperature of about 600°K was produced by a commercial oven; temperature homogeneity along the tube length (maximum variation about 0.2°K) was achieved by means of additional heating coils in front of the windows.

3.2. The Spectrograph

The absorption spectrum was photoelectrically scanned by means of a Leiss prism double-monochromator with a slitwidth of about 0.1 mm , corresponding to a halfwidth of about 1.5 \AA . The apparatus profile was measured with the use of a mercury lamp containing ^{198}Hg ; the line $\lambda 2537 \text{ \AA}$ was investigated. The profile was found to be triangular, as to be expected, if the instrument is well adjusted.

3.3. The Red Wing of the Mercury Line $\lambda 2537 \text{ \AA}$

The red wing of the mercury absorption line was determined with 4 different densities of mercury vapor (corresponding to 4 different temperatures) in the tube, yielding sufficient absorption in the wing. From measurements of the self broadening of the mercury line $\lambda 2537 \text{ \AA}$ it was found, that its contribution to the absorption coefficient observed in the presence of argon, is considerably at mercury

number densities of the order of 10^{18} cm^{-3} , especially in the far red wing. The corrected absorption coefficient, reflecting the mercury-argon interaction only, was obtained by subtracting the absorption coefficient due to selfbroadening from the one measured in the presence of argon.

In order to obtain a single profile, we have plotted the reduced absorption coefficient $k(\Delta\tilde{\nu})/Nn$ (N, n number densities of the mercury and argon atoms respectively) as function of the wave number distance $\Delta\tilde{\nu}$ from the unperturbed line. Because of the great range of the absorption coefficient involved $k(\Delta\tilde{\nu})/Nn$ was plotted on a double logarithmic scale.

Figure 1 shows the final plot. In the near red wing, between 100 and about 250 cm^{-1} the observed profile follows very closely a $\Delta\tilde{\nu}^{-3/2}$ -law. This is in agreement with KUHN's⁸ earlier photographic work. Also, our absolute values of the absorption coefficient agree with those of Kuhn, within 10% . In the region beyond 250 cm^{-1} , which was not investigated by Kuhn, the slope of $k(\Delta\tilde{\nu})$ is seen to increase monotonically with $\Delta\tilde{\nu}$. Furthermore, in this region some variation of $k(\Delta\tilde{\nu})$ with the temperature is indicated. The number of experiments performed was not sufficient, however, to confirm the effect.

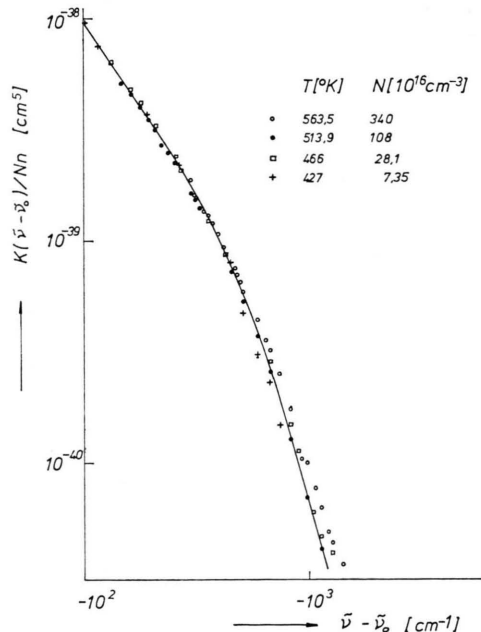


Fig. 1. The red wing of the mercury absorption line $\lambda 2537 \text{ \AA}$ broadened by argon at $1.26 \times 10^{19} \text{ cm}^{-3}$ argon number density. Measurements at 4 different mercury number densities, corresponding to 4 different temperatures. The full curve shows the shape measured at $T = 513.9^\circ \text{K}$.

4. The Interpretation of the Results

4.1. The Splitting of the $^3\text{P}_1$ -State of the Mercury Atom

For the interpretation of the experimental results it is necessary to consider the splitting of the $^3\text{P}_1$ state of the mercury atom into two substates with space quantum numbers $m_J = 0$ and ± 1 . To these substates there correspond two distinct molecular states of the diatomic system, composed of a mercury atom in its $^3\text{P}_1$ state and an argon atom in its $^1\text{S}_0$ ground state, and which are labeled, in the notation of HERZBERG⁹, $\text{B}^1\Sigma_0$ and $\text{A}^3\Pi_1$ respectively. The system with both atoms in the ground state is labeled $\text{X}^1\Sigma_0$. We denote the respective adiabatic potential functions by $V'_\Sigma(r)$, $V'_\Pi(r)$ and $V''_\Sigma(r)$, the difference potentials by $V_\Sigma(r) = V'_\Sigma(r) - V''_\Sigma(r)$ and $V_\Pi(r) = V'_\Pi(r) - V''_\Sigma(r)$.

In order to obtain information on the two difference potentials separately, we must require, that each wing is formed exclusively by transitions

$$\text{X}^1\Sigma_0 \rightarrow \text{A}^3\Pi_1 \quad \text{or} \quad \text{X}^1\Sigma_0 \rightarrow \text{B}^1\Sigma_0.$$

Lacking detailed knowledge of the functions $V_{\Sigma}(r)$ and $V_{\Pi}(r)$ for the system Hg—Ar, we cannot ascertain whether the exclusivity requirement is fulfilled for the red wing. From a rough knowledge of the relative magnitudes of the attractive and repulsive parts of the functions $V_{\Sigma}(r)$, $V_{\Pi}(r)$ and $V'_{\Sigma}(r)$ we may, however, conclude, that it is most likely fulfilled: Approximating the attractive parts by the Van der Waals term $\sim r^{-6}$, we obtain for $C'_{6\Sigma}$, $C'_{6\Sigma}$ and $C'_{6\Pi}$ the theoretical values¹⁰ 61.0, 61.6 and 100 eV Å⁶ respectively. With an estimated error of about 10% for each value one may conclude that $C_{6\Sigma}$ is very small or even negative and $C_{6\Pi}$ is definitely positive. The relative magnitude of the repulsive parts of the potentials may be seen from sketches of the electron probability distribution (Fig. 2): In the $X^1\Sigma_0$ -state the mercury electron

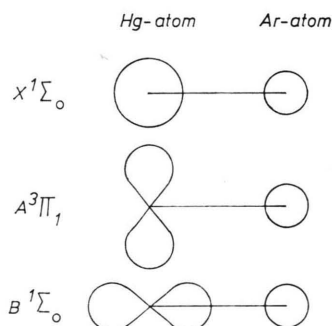


Fig. 2. Schematic representation of the electron distribution for various states of the mercury-argon diatomic system.

is mainly in a spherically symmetric $s\sigma$ -orbital. In the $B^1\Sigma_0$ -state the mercury electron is mainly in a $p\sigma$ -orbital, which overlaps the argon atom at larger internuclear distances than the $s\sigma$ -orbital. In the $A^3\Pi_1$ -state, the mercury electronic wave function has $p\pi$ -character with a node along the internuclear axis, and the argon atom can approach the mercury atom rather closely before the repulsive interaction dominates. It follows, that the $B^1\Sigma_0$ -state is the most repulsive and the $A^3\Pi_1$ -state the most attractive state. More quantitatively, if the repulsive part of the potentials is approximated by the term $\sim r^{-12}$, it follows, that $C'_{12\Sigma} > C'_{12\Sigma} > C'_{12\Pi}$. Consequently $C_{12\Sigma} = C'_{12\Sigma} - C'_{12\Sigma} > 0$ and $C_{12\Pi} = C'_{12\Pi} - C'_{12\Sigma} < 0$. In other words, the difference potential $V_{\Sigma}(r)$ will be positive and the difference potential $V_{\Pi}(r)$ will be negative over the whole r -range under investigation. We conclude, that the red wing of the mercury line λ 2537 Å perturbed by argon is exclusively formed by transitions $X^1\Sigma_0 \rightarrow A^3\Pi_1$.

4.2. The Determination of the Functions

$$V_{\Pi}(r) \text{ and } V'_{\Pi}(r)$$

Since the measured $k(\Delta\tilde{\nu})$ does not even display an inflection point, it seems very unlikely, that the strong decrease of $k(\Delta\tilde{\nu})$ beyond 250 cm⁻¹ is associated with a possible turning point of the difference potential. Also, according to Sect. 4.1, the occurrence of such a turning of $V_{\Pi}(r)$ in the range $V_{\Pi}(r) < 0$ would be very improbable. We rather assume, that this feature of $k(\Delta\tilde{\nu})$ is mainly due to the rapid increase of the ground state potential $V'_{\Sigma}(r)$ in the range $r < \sigma''$.

The evaluation of $V_{\Pi}(r)$ and $V'_{\Pi}(r)$ from $k(\Delta\tilde{\nu})$ is performed on the basis of Eq. (3), where the intensity is related to the absorption coefficient by

$$k(\Delta\nu) d(\Delta\nu) = C I(\Delta\nu) d(\Delta\nu), \\ C = N^* f(\pi e^2/mc), \quad (8)$$

(N^* effective number density of absorbing atoms, f oscillator strength of the line, e , m charge and mass of the electron, c light velocity).

Since the state $A^3\Pi_1$ has the statistical weight 2 compared with 3 for the states $A^3\Pi_1$ and $B^1\Sigma_0$ together, the effective number density N^* of mercury atoms involved in transitions $X^1\Sigma_0 \rightarrow A^3\Pi_1$ is $\frac{2}{3}N$. For the oscillator-strength of the mercury line the value 0.0255 was taken. The probability distribution $W(r)$ was calculated from (5), (6) and (7) with the use of the potential $V'_{\Sigma}(r)$ for the ground state, as calculated by HELLER¹¹ from spectroscopic data:

$$V'_{\Sigma}(r) = A \exp\{-r/\rho\} - \frac{C_6}{r^6} - \frac{C_8}{r^8} - \frac{C_{10}}{r^{10}} - \frac{C_{12}}{r^{12}}. \quad (9)$$

For $k(\Delta\tilde{\nu})$ the function measured at $T = 513.9^\circ\text{K}$ was used. For $\Delta\tilde{\nu}_0$ we have chosen the value 50 cm⁻¹ within the validity range of the $\Delta\tilde{\nu}^{-3/2}$ law, observed by KUHN⁸, which is characteristic for quasistatic interaction. With this value for $\Delta\tilde{\nu}_0$ we obtained, by numerical integration of (4), for r_0 the value 3.60 Å. The difference potential $V_{\Pi}(r)$ is then obtained from (3). From $V_{\Pi}(r)$ and $V'_{\Sigma}(r)$, given by (9), one obtains $V'_{\Pi}(r)$. These functions are plotted in Figure 3.

5. Discussion

5.1. The Potential Functions

We denote by $r'_{m\Sigma}$, $r'_{m\Pi}$ and ϵ'_{Σ} , ϵ'_{Π} the positions of the minima and the depths of the wells of the po-

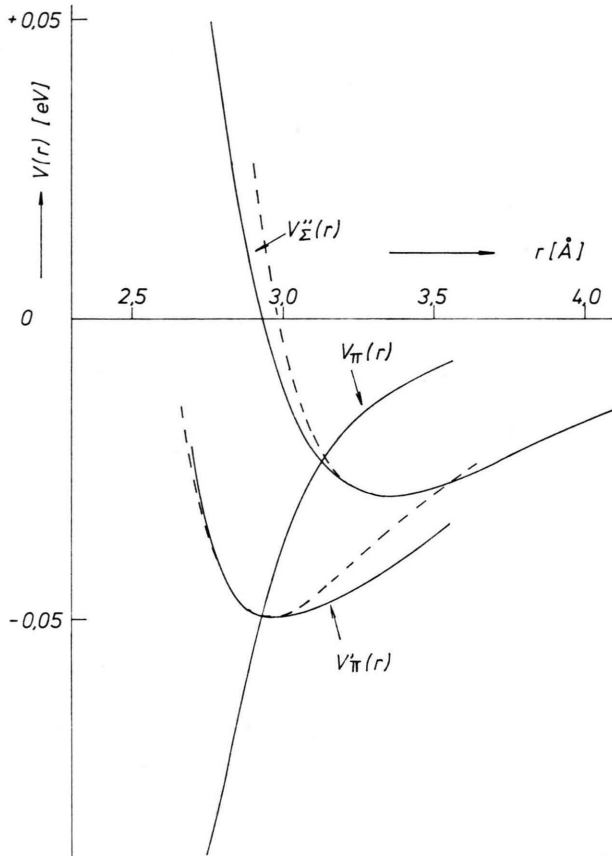


Fig. 3. Potential curves for the mercury-argon diatomic system; ——— from spectroscopic data; - - - - - adjusted Lennard-Jones-potential.

tential functions $V_{\Sigma}''(r)$ and $V_{\Pi}'(r)$, respectively. Comparing the functions, it is seen from Fig. 3, that $r_{m\Sigma}'' > r_{m\Pi}'$ and $\epsilon_{\Sigma}'' < \epsilon_{\Pi}'$. This result was to be expected from the considerations in Sect. 4.1. Also, the same behaviour is displayed by theoretical potential functions for the systems alkali-rare gas in comparable states, reported by BAYLIS¹².

It is of interest, to compare the potential function $V_{\Pi}'(r)$, obtained from the above method, with an adjusted model function of the L.-J.-(12,6)-type. The adjustment is made by requiring, that the positions of the minima and the depths of both potentials coincide. We then find, that only in the region $r < r_{m\Pi}'$ the L.-J.-potential is a good approximation to the true potential function, whereas in the range $r > r_{m\Pi}'$ the potentials deviate considerably. This deviation certainly reflects to some degree the difference between the Van der Waals constant $C'_{6\Pi L.J.} = 65 \text{ eV } \text{\AA}^6$ of the adjusted L.-J.-potential and the theoretical value $C'_{6\Pi \text{ theor}} = 100 \text{ eV } \text{\AA}^6$.

Next we compare the difference potential $V_{\Pi}(r)$ obtained from the wing with the L.-J.-potential $V(r)_{LJ}$ obtained from the core of the line. For this purpose we first approximate $V_{\Pi}(r)$ by a L.-J.-potential $V_{\Pi}(r)_{LJ}$ such, that the values of the functions at two different r coincide. We choose

$$V_{\Pi}(r_1 = 3.5 \text{ \AA}) = -0.0083 \text{ eV}$$

and

$$V_{\Pi}(r_2 = 3.0 \text{ \AA}) = -0.038 \text{ eV}$$

and obtain

$$C_{6\Pi} = 7.25 \text{ eV } \text{\AA}^6 \text{ and } C_{12\Pi} = -5.15 \times 10^4 \text{ eV } \text{\AA}^{12}.$$

On the other hand we may derive C_6 and C_{12} for $V(r)_{LJ}$ from halfwidth and shift¹³ by means of a method described elsewhere². Assuming $C_{12} > 0$, one finds from this method $C_6 = 1.94 \times 10^2 \text{ eV } \text{\AA}^6$ and $C_{12} = 1.88 \times 10^8 \text{ eV } \text{\AA}^{12}$. This potential has its minimum at 11.2 \AA and a depth of $5.0 \times 10^{-5} \text{ eV}$.

Comparing these data, we have to consider, that the values for C_6 and C_{12} are some kind of average values of the corresponding constants of $V_{\Sigma}(r)$ and $V_{\Pi}(r)$. The large value of C_{12} may be understood, if one assumes, that $V_{\Sigma}(r)$ is strongly repulsive. This assumption is in line with that made in Sect. 4.1. On the other hand, the C_6 -value is by a factor of about 25 larger than $C_{6\Pi}$. This cannot be understood, since $C_{6\Sigma}$ should be very small. We thus come to the conclusion, that the L.-J.-potential, obtained from the line core, is in our case incapable to describe the red wing of the line. The very shallow minimum at large internuclear distance in the difference potential $V(r)_{LJ}$ strongly indicates, that the potential $V_{\Pi}'(r)$ for the upper state consists of two wells: a deep one at small r and a flat one at large r . Potentials of this type have been predicted by BAYLIS¹² for excited states of various systems alkali atom-rare gas.

5.2. The Theoretical Assumptions of the Method

The resulting potential curves have been obtained on the basis of the one-particle approximation of the quasi static theory. We therefore have to discuss the validity of the assumptions made in this theory (Sect. 2.1).

The influence of the motion of the atoms on the quasi-static intensity distribution still remains to be investigated quantitatively. It is well known¹, that this influence may become considerable at the frequency corresponding to the turning point in the difference potential, where the quasistatic intensity

changes rapidly. It is not expected to be very large, however, in the present case, where the intensity change is comparatively small.

The one-particle assumption is well fulfilled: the exponential term in (5) deviates from unity only by about 0.003 at $n = 1.26 \times 10^{19} \text{ cm}^{-3}$, if the integration is extended from 0 to 3.60 Å.

It might not be excluded, that there is some change of the oscillator strength of the mercury λ 2537 Å line with the internuclear separation between mercury and argon atom. SCHULLER and OKSENGORN¹⁴ have pointed out recently, that for intercombination lines the transition moment may be increased considerably due to interaction between instantaneous dipoles of the active and perturbing atom. On the other hand, STRIJLAND and NANASSY¹⁵ have found experimentally, that the f -value of the mercury line λ 2537 Å perturbed by argon does not change up to an argon number density of about $1.6 \times 10^{22} \text{ cm}^{-3}$, corresponding to an average internuclear distance of 2.45 Å.

5.3. The Temperature Dependence of $k(\Delta\tilde{\nu})$

For $\Delta\tilde{\nu} > 400 \text{ cm}^{-1}$, corresponding to $r < \sigma''$, the slope of $k(\Delta\tilde{\nu})$ is predicted to decrease with the temperature, due to the Boltzmann factor in the probability distribution (5). From Fig. 1 it can be seen, that a temperature effect is indicated. The limited number of experiments performed and the small temperature range investigated, however, did not allow so far to verify the above prediction experimentally. The most severe test of the interpretation of the present results will be to examine, whether the intensity distribution in the quasistatic wings can be described by the same difference potential at largely differing temperatures. Measurements of the temperature dependence of the line wings are under preparation in our laboratories.

Acknowledgements

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