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CONTINUED FACTORIZATION METHOD FOR UPPER AND LOWER BOUNDS ON THE DYNAMIC POLARIZABILITY

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A continued factorization method is introduced to establish upper and lower bounds on the dynamic polarizability of both real and imaginary frequencies. It converges rapidly and has a wider frequency range of applicability than other methods.

Dynamic polarizability of real frequency is directly related to the index of refraction and Verdet constant; dynamic polarizability of imaginary frequency is intimately connected with the dispersion energy.¹ Since direct calculation is difficult, feverish search has been made recently for the determination of upper and lower bounds to this quantity.²⁻⁵

In this Letter we wish to introduce a continued factorization method which can be used to establish the bounds simply and directly. It converges very rapidly and has a wider frequency range of applicability than other methods.

The dynamic polarizability α is defined as (in a.u.)

$$\alpha(\omega) = \sum_{n=1}^{\infty} \frac{f_n}{\epsilon_n^2 - \omega^2}, \quad (1)$$

where f_n is the oscillator strength for the dipole transition from the ground state to the excited state n and ϵ_n is the associated transition energy. The summation is understood to include the integration over the continuum.

Expanding into the power series, we have the

$$\alpha(\omega) = \frac{\epsilon_1^2 \alpha_0}{\epsilon_1^2 - \omega^2} - \frac{\omega^2}{\epsilon_1^2 - \omega^2} \left\{ \frac{\epsilon_2^2 \alpha_1}{\epsilon_2^2 - \omega^2} - \frac{\omega^2}{\epsilon_2^2 - \omega^2} \left[\frac{\epsilon_3^2 \alpha_2}{\epsilon_3^2 - \omega^2} - \frac{\omega^2}{\epsilon_3^2 - \omega^2} \left(\dots \frac{\omega^2}{\epsilon_m^2 - \omega^2} \sum \frac{f_n^m}{\epsilon_n^2 - \omega^2} \right) \dots \right] \right\}, \quad (4)$$

so-called Cauchy series

$$\alpha(\omega) = \sum_{k=0}^{\infty} \mu_k (\omega^2)^k. \quad (2)$$

The first few Cauchy moments μ_k can be determined either experimentally or theoretically.¹ Obviously μ_0 is just the static polarizability. The convergence of Cauchy series is usually very poor in the extrapolated region. It is convenient to define

$$\alpha_n = \sum_{k=0}^n a_k \mu_k,$$

$$a_0 = 1,$$

$$a_1 = - \sum_{i=1}^{n'} \epsilon_i^2,$$

$$a_2 = \frac{1}{2!} \sum_{i,j=1}^{n'} \epsilon_i^2 \epsilon_j^2,$$

$$a_3 = - \frac{1}{3!} \sum_{i,j,k=1}^{n'} \epsilon_i^2 \epsilon_j^2 \epsilon_k^2, \quad (3)$$

etc., where \sum' means the summation does not include terms with two identical indices. With these quantities, we can express the dynamic polarizability $\alpha(\omega)$ in the form of continued factorization,

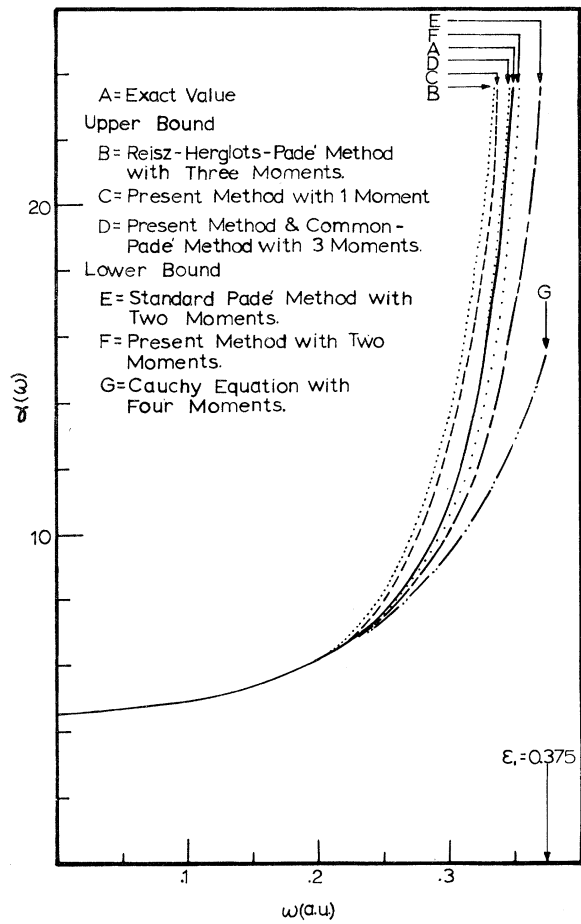


FIG. 1. Dynamic polarizability of real frequency for the hydrogen atom.

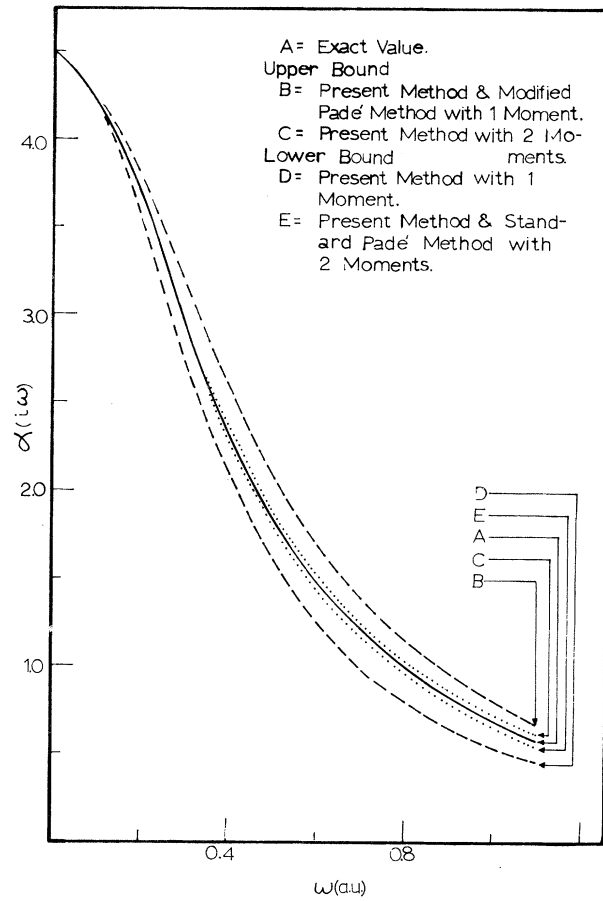


FIG. 2. Dynamic polarizability of imaginary frequency for the hydrogen atom.

where

$$f_n^m = \frac{(\epsilon_n^2 - \epsilon_1^2)(\epsilon_n^2 - \epsilon_2^2) \cdots (\epsilon_n^2 - \epsilon_m^2)}{\epsilon_n^{2m}} f_n. \tag{5}$$

If we terminate the series after the n th term we have the n th order approximation. Thus the first-order approximation is

$$\alpha^{(1)}(\omega) = \frac{\epsilon_1^2 \alpha_0}{\epsilon_1^2 - \omega^2} \tag{6}$$

and the n th-order approximation is

$$\alpha^{(n)}(\omega) = \alpha^{(n-1)} + (-1)^{n-1} \left[\prod_{i=1}^n \frac{1}{\epsilon_i^2 - \omega^2} \right] \omega^{2(n-1)} \epsilon_n^2 \alpha_{n-1}. \tag{7}$$

By examining the remainder, we can establish the bounding properties of all orders of approximation. It can be shown that $\alpha^{(n)}(\omega)$ is an upper bound to $\alpha(\omega)$ in the frequency range $\epsilon_{k-1} < \omega < \epsilon_k$ and a lower bound in the range $\epsilon_k < \omega < \epsilon_{k+1}$, where $k = n, n-2, n-4, \dots$. All previous bounding methods are limited to the normal dispersion region ($\omega < \epsilon_1$) while the present method can give the bounds between the excitation energies as well. As an illustration we present the $\alpha(\omega)$ of the hydrogen atom and various bounds in Fig. 1. As a lower bound, the present method converges much faster than the standard Padé method^{3,6}; as an upper bound, it is superior to the Reisz-Herglotz-Gronwall-Padé method^{1,6} and converges at about the same rate as the Common-Padé method.^{3,7} The lowest-order Common-Padé

bound requires three Cauchy moments while with the present method the upper bound can be constructed with a single moment (the static polarizability).

For imaginary frequency, we can use the same form of continued factorization of Eq. (4). One can show, by examining the remainder, that $\alpha^{(n)}(i\omega)$ is a lower bound to $\alpha(i\omega)$ for all orders of n . To construct an upper bound with the present method, we can make use of the total number of electrons Z in the atom. If we define

$$Z_n = Z + \sum_{k=1}^n a_k \mu_{k-1}, \quad (8)$$

we can show that

$$\beta^{(n)}(i\omega) = \frac{\epsilon_1^2 \alpha_0}{\epsilon_1^2 + \omega^2} + \frac{\omega^2}{\epsilon_1^2 + \omega^2} \left\{ \frac{\epsilon \alpha}{\epsilon_2^2 + \omega^2} + \frac{\omega}{\epsilon_2^2 + \omega^2} \left[\frac{\epsilon_3^2 \alpha_2}{\epsilon_3^2 + \omega^2} + \frac{\omega^2}{\epsilon_3^2 + \omega^2} \left(\cdots + \frac{\omega^2}{\epsilon_n^2 + \omega^2} \cdot \frac{Z_n \alpha_n}{Z_n + \alpha_n \omega^2} \right) \cdots \right] \right\} \quad (9)$$

is an upper bound to $\alpha(i\omega)$ for all n . This is because

$$\frac{Z_n \alpha_n}{Z_n + \alpha_n \omega^2} \geq \sum_j \frac{f_j^n}{\epsilon_j^2 + \omega^2}, \quad (10)$$

which corresponds to the two-point Slater-Kirkwood-Padé inequality.⁵ These bounds are illustrated in Fig. 2 for the hydrogen atom. The convergence rate is about the same as the standard Padé approximant. To tighten either the upper or the lower bound, the Padé method requires two Cauchy moments at a time whereas with the present method we need only one Cauchy moment for each step.

¹For a recent review, see P. M. Langhoff and M. Karplus, in "The Padé Approximant in Theoretical Physics," edited by G. A. Baker, Jr., and J. L. Gammel (Academic Press, Inc., New York, to be published).

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REMEASUREMENT OF ΔE - \mathcal{S} IN ATOMIC HYDROGEN*

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The $2^2S_{1/2}$ - $2^2P_{3/2}$ separation ΔE - \mathcal{S} in hydrogen has been measured by a microwave atomic-beam technique. The result obtained from four independent transitions is ΔE - $\mathcal{S} = 9911.173 \pm 0.042$ MHz. We combine this result with the recent determination of the Lamb shift \mathcal{S} by Robiscoe to obtain a value for the fine-structure splitting ΔE and a value for the fine-structure constant $\alpha^{-1} = 137.0358(5)$.

We have measured ΔE - \mathcal{S} , the splitting between the states $2^2S_{1/2}$ and $2^2P_{3/2}$ in atomic hydrogen (Fig. 1). The experiment is modeled on the original work of Lamb and co-workers.¹ A beam of hydrogen atoms is produced with a single hyperfine component of the $2^2S_{1/2}(m_J = -\frac{1}{2})$ metastable level. The beam is subjected to an rf electric field in a magnetic field of ≈ 400 G oriented parallel to the beam axis. Electric dipole transitions

are thereby induced to the b or d levels in the $2^2P_{3/2}$ state (see Fig. 1). We have measured four independent transitions. Table I shows the initial and final states along with the frequency and magnetic field for each.

During a data run, the frequency of the electric field is held constant to 1 ppm, and the magnetic field is swept through the resonance line. A typical experimental resonance is plotted in Fig. 2