

## Discrete basis representation of Ursell operators

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The inverse Laplace transform of the two- and three-particle Ursell operators are shown to be related to scattering kernels. For a three-particle system the kernel is identical to Faddeev's connected kernel. For well-behaved potentials, these kernels are compact with the consequence that they have a discrete spectrum and can thus be expressed in terms of discrete spectral representations. This leads to a method for the direct computation of Ursell operators and the corresponding cluster integrals. [S1063-651X(96)02709-2]

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### I. INTRODUCTION

This paper explores the formal relation between the connected kernel of Faddeev [1] and the three-particle Ursell operator of statistical mechanics [2,3]. The convergence properties of the Faddeev kernel thus leads to an alternate method of evaluating the quantum mechanical cluster integrals of equilibrium statistical mechanics. While the present paper is restricted to three-particle systems and Boltzmann statistics, it is expected that this correspondence between connected scattering kernels and Ursell operators can be generalized to the higher order Ursell operators and to quantum statistics.

The main purpose of such a formulation is to provide a practical method for the quantum mechanical evaluation of the equation of state of a nonideal gas system whose standard starting point is the cluster expansion of the grand partition function, a classic approach developed by Ursell [2] and Mayer [4] and later generalized to quantum mechanics by Kahn and Uhlenbeck [5]. Alternately, Dashen, Ma, and Bernstein [6] used a Feynman-Dyson expansion for the grand partition function and their use of Feynman diagrams allows a separation between a "dynamical part" and a "statistical part." A modern scattering theory formulation of a monomer-dimer mixture has been given by Osborn [7]. His method relies on the asymptotic completeness theorem of the multichannel scattering theory. An elegant two-Hilbert space formalism for the multispecies fugacity expansion has been provided by Hoffman and Evans [8]. Their formalism is based on the method used in a rigorous reactive quantum kinetic theory developed by Hoffman, Kouri, and their co-workers [9].

The first quantum mechanical calculation of the second virial coefficient was due to Beth and Uhlenbeck [10], expressed in terms of scattering phase shifts. Since the phase shifts are related to the  $S$  matrix, the Jost function, and the on-shell  $T$  matrix, various alternative expressions of this result have been reported [11–13]. Smith [14] relates the energy derivative of the phase shift to the time delay and generalized this to time delay matrices for general inelastic collisions. Thus the pair particle Ursell operator can be expressed in terms of a time delay. Higher order Ursell operators can also be expressed in terms of a time delay formalism [15], for example, the general  $S$ -matrix formulation of equilibrium statistical mechanics [6] is basically in terms of the

time delay. The most elegant formalism for the virial coefficients using the time delay method has been given by Osborn [7]. When bound states exist, his work expresses the correlations of dimer-dimer, monomer-dimer, and monomer-monomer in terms of the corresponding time delays on an equal footing. Despite the various developments, the evaluation of the second virial coefficient is nontrivial except for a limited class of simple potentials [16]. It is even more difficult to evaluate higher order virials because, for example, these depend on evaluating the full  $S$  matrix for all energies. Therefore reliable alternate numerical methods are indispensable for solving realistic problems. The Hilbert-Schmidt method is an extremely powerful technique for solving quantum mechanical two-particle scattering problems [17–19], however, it has rarely been used in formal statistical mechanics. Necessarily, calculating higher order virial coefficients rests on three- or more body quantum scattering theory, where the Faddeev-type analysis is obligatory. In this paper the Faddeev analysis is employed to formulate a Hilbert-Schmidt representation of the three-particle Ursell operator.

The resulting formalism is in a form ready for direct computation. An example of such a computation, for the pair-particle Ursell operator, was considered [20] earlier.

A discrete basis representation of Ursell operators is presented in Sec. II. Ursell operators are first expressed in terms of various resolvents by a Laplace transform [21]. Operator theory implies that certain kernel operators are compact so that a discrete basis representation can be introduced. In the case of three particles, the Faddeev analysis is utilized to obtain a discrete basis expansion of the corresponding Ursell operator. A brief discussion is given in Sec. III.

### II. DISCRETE BASIS REPRESENTATION OF URSELL OPERATORS

In this section a discrete basis representation for the two- and three-particle Ursell operators is formulated. This section is divided into two parts, the first dealing with the two-particle Ursell operator and the second with the three-particle Ursell operator.

#### A. Two-particle Ursell operator

In thermal equilibrium the correlations between two particles is given by the Ursell operator

$$U^{(2)} = e^{-\beta H^{(2)}} - e^{-\beta H_0^{(2)}}. \quad (1)$$

Here the full and free Hamiltonians  $H^{(2)}$  and  $H_0^{(2)}$  for relative motion are self-adjoint operators acting on the (relative motion) pair Hilbert space  $\mathfrak{H}^{(2)}$  with both their spectra being absolutely continuous on the positive real axis and there is the added possibility that the spectrum of  $H^{(2)}$  is also singular on the negative real axis, corresponding to a finite number of bound states. As long as the potential  $V \equiv H^{(2)} - H_0^{(2)}$  is well behaved,  $U^{(2)}$  is a trace class operator [22,23]

$$\text{Tr}|U^{(2)}| < \infty. \quad (2)$$

$U^{(2)}$  is related to the interacting and free resolvent operators  $R^{(2)}$  and  $R_0^{(2)}$ ,

$$R^{(2)} \equiv \frac{1}{z - H^{(2)}}, \quad R_0^{(2)} \equiv \frac{1}{z - H_0^{(2)}}, \quad (3)$$

by the Laplace transform [21]

$$U^{(2)} = \frac{1}{2\pi i} \oint_C e^{-\beta z} \mathcal{R}^{(2)}(z) dz, \quad (4)$$

expressed as a contour  $C$  counterclockwise encircling the spectrum of  $H^{(2)}$ , of the difference of the resolvent operators

$$\mathcal{R}^{(2)}(z) \equiv R^{(2)} - R_0^{(2)} = \frac{1}{z - H^{(2)}} - \frac{1}{z - H_0^{(2)}}. \quad (5)$$

The corresponding resolvents  $(z - H^{(2)})^{-1}$  and  $(z - H_0^{(2)})^{-1}$  are bounded operators on  $\mathfrak{H}^{(2)}$  for all complex  $z$  except on the spectra of their respective Hamiltonians. As a consequence, the difference in resolvents  $\mathcal{R}^{(2)}(z)$  is defined on the complex  $z$  plane excluding the spectrum of  $H^{(2)}$ . An advantage of  $\mathcal{R}^{(2)}$  over the individual resolvent operators  $R^{(2)}$  and  $R_0^{(2)}$  is that, for reasonable potentials, the operator  $\mathcal{R}^{(2)}$  is of trace class, whereas  $R^{(2)}$  and  $R_0^{(2)}$  are not.

It is easy to show that the difference in resolvents  $\mathcal{R}^{(2)}$  is related to the kernel

$$K^{(2)}(z) \equiv (z - H_0^{(2)})^{-1} V, \quad (6)$$

by

$$\mathcal{R}^{(2)}(z) = \left( \frac{1}{1 - K^{(2)}(z)} \right) K^{(2)}(z) \left( \frac{1}{z - H_0^{(2)}} \right). \quad (7)$$

Moreover the kernel  $K^{(2)}(z)$  is, for  $\sqrt{2\mu z} = x + iy$  not on the real axis and for square integrable local potentials, a Hilbert-Schmidt class operator [24,25]. This follows from the fact that the operator  $K^{(2)}(z)^\dagger K^{(2)}(z)$  has a finite trace, namely, for a three dimensional system,

$$\begin{aligned} \text{Tr} K^{(2)}(z)^\dagger K^{(2)}(z) &= \int \langle \mathbf{p} | V^2 | \mathbf{p} \rangle \\ &\times \frac{4\mu^2}{[(x-p)^2 + y^2][(x+p)^2 + y^2]} d\mathbf{p} < \infty, \end{aligned} \quad (8)$$

provided  $V$  is a square integrable local potential, namely,

$$\langle \mathbf{p} | V^2 | \mathbf{p} \rangle = \frac{1}{h^3} \int V(\mathbf{r})^2 d\mathbf{r} \quad (9)$$

is finite. As a consequence,  $K^{(2)}(z)$  is a compact operator and its spectrum discrete [25], having eigenvalues  $\eta_n(z)$ , and a set of biorthonormal right  $|\zeta_n(z)\rangle$  and left  $\langle \xi_n(z)|$  eigenvectors indexed by  $n$ ,

$$K^{(2)}(z) |\zeta_n(z)\rangle = \eta_n(z) |\zeta_n(z)\rangle, \quad (10)$$

$$\langle \xi_n(z) | K^{(2)}(z) = \langle \xi_n(z) | \eta_n(z), \quad (11)$$

$$\langle \xi_m(z) | \zeta_n(z) \rangle = \delta_{mn}, \quad (12)$$

with the eigenvalues satisfying

$$\sum_n |\eta_n(z)|^2 < \infty. \quad (13)$$

The proof of the compactness of  $K^{(2)}(z)$  can be extended to the real axis, see, e.g., Ref. [26], by showing that the symmetrized kernel  $V^{1/2}(z - H_0^{(2)})^{-1} V^{1/2}$  is a Hilbert-Schmidt operator.

It follows that  $\mathcal{R}^{(2)}(z)$  also has a representation in terms of a discrete basis

$$\mathcal{R}^{(2)}(z) = \sum_n |\zeta_n(z)\rangle \frac{\eta_n(z)}{1 - \eta_n(z)} \langle \xi_n(z) | \frac{1}{z - H_0^{(2)}}. \quad (14)$$

Thus one arrives at a discrete basis representation for the pair Ursell operator

$$U^{(2)} = \sum_n \frac{1}{2\pi i} \oint_C e^{-\beta z} |\zeta_n(z)\rangle \frac{\eta_n(z)}{1 - \eta_n(z)} \langle \xi_n(z) | \frac{1}{z - H_0^{(2)}} dz. \quad (15)$$

### B. Three-particle Ursell operator

At thermal equilibrium three-particle correlations are described by the three-body Ursell operator

$$U^{(3)} = e^{-\beta H^{(3)}} - e^{-\beta H_0^{(3)}} - \sum_\alpha (e^{-\beta H_\alpha^{(3)}} - e^{-\beta H_0^{(3)}}). \quad (16)$$

Here the full three-body Hamiltonian for relative motion is defined as

$$H^{(3)} \equiv H_0^{(3)} + V^{(3)}. \quad (17)$$

$H_0^{(3)}$  is the kinetic energy operator for the relative motion of three particles and  $V^{(3)}$  is the pairwise additive potential

$$V^{(3)} \equiv \sum_\alpha V_\alpha, \quad (18)$$

given in terms of the pair potentials

$$V_1 \equiv V_{23}, \quad V_2 \equiv V_{13}, \quad V_3 \equiv V_{12} \quad (19)$$

labeled in a convenient (and standard) manner for the subsequent discussion. Arrangement channel Hamiltonians  $H_\alpha^{(3)}$  are defined according to

$$H_\alpha^{(3)} \equiv H_0^{(3)} + V_\alpha. \quad (20)$$

The three-body Hamiltonians are self-adjoint operators acting on the relative motion Hilbert space  $\mathfrak{H}^{(3)}$  [the center of mass motion is assumed to have been separated off]. The spectrum of  $H^{(3)}$  is absolutely continuous on the positive real axis corresponding to three-particle scattering, and possibly on the part of the negative real axis corresponding to two-fragment scattering subsystems [with one of the pair of particles in a bound state if such exist]. As well, the spectrum of  $H^{(3)}$  can also be singular to the left of the two-particle threshold, corresponding to three-particle bound states.

The full three-body resolvent operator is defined as

$$R^{(3)}(z) = \frac{1}{z - H^{(3)}}, \quad (21)$$

where  $z$  is a parameter in the complex energy plane. As a function of  $z$ ,  $R^{(3)}(z)$  is well defined in the whole complex energy plane excluding the spectrum of the Hamiltonian  $H^{(3)}$ , specifically off the real axis and to the left (more negative than) the lowest bound state energy. Similarly, the three-body channel resolvent operators are defined as

$$R_\alpha^{(3)}(z) = \frac{1}{z - H_\alpha^{(3)}}. \quad (22)$$

In terms of these resolvent operators and the Laplace transform, the three-body Ursell operator can be expressed as

$$U^{(3)} = \frac{1}{2\pi i} \oint_C e^{-\beta z} \mathcal{R}^{(3)}(z) dz, \quad (23)$$

where the contour  $C$  encircles the spectra of the full three-body Hamiltonian in a counterclockwise manner and the quantity  $\mathcal{R}^{(3)}(z)$  is the combination of resolvents

$$\mathcal{R}^{(3)}(z) \equiv R^{(3)}(z) - R_0^{(3)}(z) - \sum_\alpha [R_\alpha^{(3)}(z) - R_0^{(3)}(z)]. \quad (24)$$

$\mathcal{R}^{(3)}$  has been shown to be a connected operator [27]. Actually  $\mathcal{R}^{(3)}$  is also related to the Faddeev kernel of the three-body transition operator, which is a compact operator, a necessary functional property for the validity of a representation in terms of a discrete basis. The Faddeev technique of three-body quantum scattering is utilized in the following to establish this relation between the operator  $\mathcal{R}^{(3)}(z)$  and the Faddeev kernel.

Faddeev found that the three-body (on-shell) Lippmann-Schwinger equation [28] does not have a unique solution because the homogeneous equations admit two-body scattering state solutions. The Lippmann-Schwinger equation is thus equivalent to the three-body Schrödinger equation only for energies below the lowest two-particle threshold. At higher energies the kernel of the Lippmann-Schwinger equation becomes noncompact so that the equation is no longer of

the Fredholm type and the standard theory of integral equations cannot be applied. In spite of these difficulties, a three-body transition operator can still be defined in a form analogous to the two-body transition operator, namely,

$$T^{(3)}(z) \equiv V^{(3)} + V^{(3)}R^{(3)}(z)V^{(3)}. \quad (25)$$

However,  $T^{(3)}(z)$  does not give the scattering cross section in the same manner as does the two-body transition operator. The three-body resolvent operator can be written in the following alternate ways:

$$R^{(3)}(z) = R_\alpha^{(3)}(z) + R_\alpha^{(3)}(z)V^\alpha R^{(3)}(z) \quad (26)$$

$$= R_\alpha^{(3)}(z) + R^{(3)}(z)V^\alpha R_\alpha^{(3)}(z) \quad (27)$$

$$= R_0^{(3)}(z) + R_0^{(3)}(z)T^{(3)}(z)R_0^{(3)}(z), \quad (28)$$

where

$$V^\alpha \equiv V^{(3)} - V_\alpha. \quad (29)$$

$R^{(3)}(z)$  is more singular than the transition operator  $T^{(3)}(z)$  because of the presence of  $R_0^{(3)}(z)$ . In graph theory language [29], Eq. (26) has disconnected diagrams which give rise to  $\delta$  functions in momentum representation.

Faddeev separated the transition operator into three parts  $T^{(3)} = \sum_\alpha T_\alpha^{(3)}(z)$ , satisfying the set of equations

$$T_\alpha^{(3)}(z) \equiv V_\alpha + V_\alpha R_0^{(3)}(z)T^{(3)} = t_\alpha^{(3)}(z) + \sum_\beta (1 - \delta_{\alpha,\beta}) \\ \times t_\alpha^{(3)}(z)R_0^{(3)}(z)T_\beta^{(3)}(z), \quad (30)$$

where  $t_\alpha^{(3)}(z)$  is a two-body transition operator in the three-particle space

$$t_\alpha^{(3)}(z) \equiv V_\alpha + V_\alpha R_\alpha^{(3)}(z)V_\alpha \quad (31)$$

$$= V_\alpha + V_\alpha R_0^{(3)}(z)t_\alpha^{(3)}(z) \quad (32)$$

$$= [1 - V_\alpha R_0^{(3)}(z)]^{-1} V_\alpha. \quad (33)$$

In an analogous manner the resolvent operator can be divided up according to

$$R^{(3)}(z) - R_0^{(3)}(z) = \sum_\alpha R_0^{(3)}(z)T_\alpha^{(3)}(z)R_0^{(3)}(z) \quad (34)$$

$$= \sum_\alpha G_\alpha^{(3)}(z). \quad (35)$$

The  $G_\alpha^{(3)}(z)$  are Faddeev's resolvent operators

$$G_\alpha^{(3)}(z) \equiv R_0^{(3)}(z)T_\alpha^{(3)}(z)R_0^{(3)}(z) \quad (36)$$

$$= R_0^{(3)}(z)t_\alpha^{(3)}(z)R_0^{(3)}(z) + \sum_\beta K_{\alpha\beta}^{(3)}(z)G_\beta^{(3)}(z) \quad (37)$$

$$= R_\alpha^{(3)}(z) - R_0^{(3)}(z) + \sum_\beta K_{\alpha\beta}^{(3)}(z)G_\beta^{(3)}(z), \quad (38)$$

with kernel  $K_{\alpha\beta}^{(3)}(z)$  defined as

$$K_{\alpha\beta}^{(3)}(z) = R_0^{(3)}(z) t_\alpha^{(3)}(z) (1 - \delta_{\alpha,\beta}). \quad (39)$$

The last form for  $G_\alpha^{(3)}$  Eq. (38), is obtained from Eq. (37) using the  $R_\alpha^{(3)}$  analog of Eq. (34), there being only one potential in this case. Using Eqs. (24), (35), and (38),  $\mathcal{R}^{(3)}(z)$  can be written as

$$\mathcal{R}^{(3)}(z) = \sum_\alpha \sum_\beta K_{\alpha\beta}^{(3)}(z) G_\beta^{(3)}(z). \quad (40)$$

On interpreting  $G_\alpha^{(3)}(z)$  and  $t_\alpha^{(3)}(z)$  as components of the channel space vectors  $\mathbf{G}$  and  $\mathbf{t}$ , together with  $K_{\alpha\beta}^{(3)}(z)$  being a matrix  $\mathbf{K}$ , this in addition to these quantities being operators on the Hilbert space of relative motion for the three particles, then Eq. (37) can be written as the matrix equation

$$\mathbf{G} = R_0^{(3)} \mathbf{t} R_0^{(3)} + \mathbf{K} \mathbf{G}, \quad (41)$$

whose formal solution is

$$\mathbf{G} = (\mathbf{1} - \mathbf{K})^{-1} R_0^{(3)} \mathbf{t} R_0^{(3)}. \quad (42)$$

As a consequence,  $\mathcal{R}^{(3)}(z)$  is given by

$$\mathcal{R}^{(3)}(z) = \sum_\alpha [\mathbf{K} \mathbf{G}]_\alpha = \sum_{\alpha,\beta} \left( \frac{\mathbf{K}}{\mathbf{1} - \mathbf{K}} R_0^{(3)}(z) \mathbf{t} R_0^{(3)}(z) \right)_{\alpha\beta}. \quad (43)$$

The compactness of the operator  $\mathcal{R}^{(3)}(z)$  is established because the Faddeev kernel  $\mathbf{K}$  is compact. Technically this has been shown [1,19] by demonstrating that the square operator  $\mathbf{K}^2$  is an operator of Hilbert-Schmidt class, namely,

$$\sum_\alpha \text{Tr}[\mathbf{K}^{2\dagger} \mathbf{K}^2]_{\alpha\alpha} < \infty. \quad (44)$$

Necessarily this requires both the quantum trace  $\text{Tr}$  and the channel trace  $\sum_\alpha$ .

As a consequence of being compact (or completely continuous),  $\mathbf{K}$  has only a discrete spectrum. Moreover,  $[\mathbf{1} - \mathbf{K}]^{-1}$  exists [30] and is a meromorphic function of  $z$ . Thus  $\mathbf{K}$  has a biorthogonal set of left and right eigenfunctions and eigenvalues

$$\mathbf{K} |\xi_n^{(3)}(z)\rangle = \eta_n^{(3)}(z) |\xi_n^{(3)}(z)\rangle, \quad (45)$$

$$\langle \xi_n^{(3)}(z) | \mathbf{K} = \langle \xi_n^{(3)}(z) | \eta_n^{(3)}(z), \quad (46)$$

$$\langle \xi_m^{(3)}(z) | \xi_n^{(3)}(z) \rangle = \delta_{mn}. \quad (47)$$

Note that the inner product, operator, ket and bra in this development act both in the relative motion of the three particles and in the channel space. The eigenvalues  $\eta_n^{(3)}(z)$  satisfy

$$\sum_n |\eta_n^{(3)}(z)|^4 < \infty. \quad (48)$$

$\mathcal{R}^{(3)}$  thus admits the expansion

$$\begin{aligned} \mathcal{R}^{(3)}(z) &= \sum_n \sum_{\alpha,\beta} \left[ \left[ |\xi_n^{(3)}(z)\rangle \frac{\eta_n^{(3)}(z)}{1 - \eta_n^{(3)}(z)} \langle \xi_n^{(3)}(z) | \right]_{\alpha\beta} \right] \\ &\times R_0^{(3)}(z) t_\beta^{(3)}(z) R_0^{(3)}(z). \end{aligned} \quad (49)$$

This leads to the desired discrete representation for the three-particle Ursell operator

$$\begin{aligned} U^{(3)} &= \frac{1}{2\pi i} \oint_C e^{-\beta z} \sum_n \sum_{\alpha,\beta} \left[ \left[ |\xi_n^{(3)}(z)\rangle \frac{\eta_n^{(3)}(z)}{1 - \eta_n^{(3)}(z)} \langle \xi_n^{(3)}(z) | \right]_{\alpha\beta} \right] \\ &\times R_0^{(3)}(z) t_\beta^{(3)}(z) R_0^{(3)}(z) dz. \end{aligned} \quad (50)$$

### III. DISCUSSION

It is seen, for the cases that have been explored, that the connected Ursell operators  $U^{(2)}$  and  $U^{(3)}$ , have a one-to-one correspondence with Hilbert-Schmidt class operators through a Laplace transform. It is believed that this is also true for the higher order Ursell operators  $U^{(4)}$ ,  $U^{(5)}$ , etc. Therefore  $U^{(4)}$  may be closely related to the Yakubovsky equations [31]. It is well known that quantum mechanical scattering theories written in terms of integral equations for four and more particle systems are extremely complicated [31] because the kernels may be unconnected. The present study implies that it may be possible to have a systematic procedure for obtaining Hilbert-Schmidt class kernels for the resolvent operators associated with the higher order Ursell operators  $U^{(n)}$ . Furthermore, the connected graphs of the Ursell-Mayer expansion [3] may have a one-to-one correspondence with the connected diagrams of Faddeev for four and more particles.

The Hilbert-Schmidt method has been widely used in two-body quantum scattering [17–19] to evaluate both scattering wave functions and collision cross sections. The Faddeev analysis has paved the way for applying the same method to three and more particle scattering [32,33]. The method provides a powerful representation both conceptually and computationally. However, it has hardly been used in formal statistical theory, as far as the authors know. Here a general formalism for the Hilbert-Schmidt representation of low order Ursell operators has been presented. The formalism may have important applications. A first and most important application is that it provides a convenient starting point for the practical calculation of the equation of state (virial coefficients), which was pursued, for the second virial coefficient, in an earlier paper [20]. A second application is the possibility of establishing generalizations of Levinson's theorem for two- and more particle scattering [34] by the resolvent method. The conventional  $S$ -matrix phase shift does not work in the case of three-body scattering since three-body breakup collisions admit no phase-shift description.

Gibson [35], Baumgartl [11], and Reiner [36] have applied the Faddeev results to calculate the third virial coefficient. Gibson expresses the third virial coefficient in terms of Faddeev's  $T$  matrix. Baumgartl treats a part of the third virial

coefficient in terms of a two-particle scattering amplitude, whereas Reiner's result is more inclusive. Since none of these authors had considered a general Hilbert-Schmidt representation for the evaluation of the virial coefficients, the present approach introduces a different and hopefully effective means for that evaluation.

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