Formation of Atomic Tritium Clusters and Bose-Einstein Condensates

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We present an extensive study of the static and dynamic properties of systems of spin-polarized tritium atoms. In particular, we calculate the two-body $|F, m_F\rangle = |0, 0\rangle$ s-wave scattering length and show that it can be manipulated via a Feshbach resonance at a field strength of about 870 G. Such a resonance might be exploited to make and control a Bose-Einstein condensate of tritium in the $|0, 0\rangle$ state. It is further shown that the quartet tritium trimer is the only bound hydrogen isotope and that its single vibrational bound state is a Borromean state. The ground state properties of larger spin-polarized tritium clusters are also presented and compared with those of helium clusters.

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In 1976 [1], Stwalley and Nosanow suggested, based on statistical arguments, that the system of spin-polarized bosonic tritium atoms behaves "very much like ^{4}He." To the best of our knowledge, their arguments have not yet been tested by a microscopic quantum mechanical treatment. A detailed theoretical study of spin-polarized tritium systems, namely, spin-polarized atomic tritium clusters and optically pumped tritium condensates, is the objective of the present work. In this Letter, we present results for both structural and scattering properties of tritium dimers, trimers, and clusters.

Pioneering experimental studies of the lowest quartet state of spin-polarized atomic trimers have been pursued recently for sodium and potassium [2]. A study of these trimers, which were prepared on the surface of large ^{4}He clusters, revealed that three-body effects are surprisingly important [3]. We are not aware, though, of any experimental or theoretical studies of larger spin-polarized atomic cluster systems. Bosonic helium systems—i.e., liquid bulk ^{4}He, two-dimensional ^{4}He films, and finite size $^4\mathrm{He}_N$ clusters [4]—have, of course, been studied extensively. This Letter thus presents the first predictions for spin-polarized atomic clusters. In particular, we characterize spin-polarized tritium clusters [in the following denoted by $(T\uparrow)_N$] with up to $N = 40$ tritium atoms, and compare their energetic and structural properties with those of bosonic $^4\mathrm{He}_N$ clusters. We hope that this study will stimulate further experimental work. Of particular interest is the lowest quartet state of the tritium trimer, which we predict to be a Borromean or halo state, and tritium cluster formation in the presence of an external magnetic field.

We also point out the possibility for creating an optically pumped gaseous tritium condensate. Bose-Einstein condensates (BECs) are, to a very good approximation, well characterized by the two-body s-wave scattering length between two atoms. It will be shown below that the triplet two-body s-wave scattering length a_t of two tritium atoms is large and negative, implying an unstable condensate of spin-polarized tritium atoms. We find, though, that there is an unusually broad Feshbach resonance [5] for two high-field-seeking $|F, m_F\rangle = |0, 0\rangle$ tritium atoms (F denotes the total angular momentum, and m_F the magnetic quantum number of this state; $F = I + J$, where I denotes the nuclear spin and J the total electron angular momentum).

While condensation of atomic hydrogen was realized experimentally in 1998 [6], it was a difficult experiment—at least in part because of hydrogen's small triplet scattering length that limits the utility of evaporative cooling. Nevertheless, owing to hydrogen's simplicity, it remains an important species to study. For instance, properties such as the interatomic potential and spin relaxation rates can be obtained theoretically from first principles. Unfortunately, we find no Feshbach resonance at reasonable field strengths for hydrogen. Thus, the resonance for tritium may permit faster condensation of a hydrogenlike atom, and allow for the formation of a stable BEC of tritium atoms with controllable properties in an optical dipole trap [7]. Formation of such a tritium condensate should enhance the lively interplay between theory and experiment.

The behavior of atomic tritium clusters and condensates is primarily determined by the two-body interaction potential for two tritium atoms, which is identical to that for two H or D atoms, except for the isotope-dependent adiabatic correction. Since there are only two electrons, these dimers are among the few for which highly accurate *ab initio* potentials are available. In the following, we concentrate on the singlet ground state ($S = 0$, $X^1\Sigma_g^+$), and on the triplet ground state ($S = 1$, $b^3\Sigma_u^+$) of the tritium dimer.

To construct the two-body $S = 0$ and $S = 1$ Born-Oppenheimer interaction potentials for hydrogen and its heavier isotopes, highly accurate *ab initio* data for the short range part [8] that incorporate the mass-dependent adiabatic correction are connected smoothly with an analytical expression describing the long-range behavior

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[9–11]. This procedure results in six potential curves describing H_2, D_2, and T_2 in their $S = 0$ and $S = 1$ states, respectively, which are then used in the radial Schrödinger equation describing the relative motion of a particle with reduced mass $m/2$. The mass dependence of the singlet and triplet scattering lengths has been discussed extensively in the literature [12,13]. Here, we estimate the uncertainty of our two-body scattering observables by solving the radial Schrödinger equation using both the reduced atomic mass and the reduced nuclear mass for each two-body potential described above.

Figure 1 compares the $(\mathrm{T}\uparrow)_2$ potential (solid line) with the similarly shallow H_2 ground state potential, LM2M2 from Aziz and Slaman [14] (dotted line). The T↑ dimer and the He dimer potentials have a well depth of $D_e = -4.6\,\mathrm{cm}^{-1}$ and $-7.6\,\mathrm{cm}^{-1}$, respectively. Note that, on the scale shown in Fig. 1, the H↑ and D↑ dimer potentials would be indistinguishable from the T↑ dimer potential. The minimum of the $(\mathrm{T}\uparrow)_2$ potential lies at a significantly larger interparticle distance ($r_e = 7.8$ a.u.) than for the He dimer ($r_e = 5.6$ a.u.). Given the lighter mass of the tritium atom ($m = 5496.9$ a.u.) compared to that of the ^{4}He atom ($m = 7296.3$ a.u.), it is not surprising that the T↑ dimer is not bound, even though the tritium van der Waals coefficient $C_6 = 6.499$ a.u. is larger than that for He, $C_6 = 1.367$ a.u. (recall that the ^{4}He dimer binding energy is only $-9.1 \times 10^{-4}\,\mathrm{cm}^{-1}$ [15]). For comparison, the inset of Fig. 1 shows the tritium triplet potential (solid line) together with the tritium singlet potential (dashed line). The singlet curve is almost 4 orders of magnitude deeper than the triplet curve and supports 27 vibrational s-wave bound states.

In agreement with values tabulated in the literature [12], we calculate the two-body s-wave triplet scattering lengths a_t for $(\mathrm{H}\uparrow)_2$ to be $a_t = 1.33$ a.u. (1.33 a.u.), and for $(\mathrm{D}\uparrow)_2$ to be -6.89 a.u. (-6.88 a.u.), using the reduced atomic (reduced nuclear) mass. From symmetry considerations, the s-wave scattering length for $(\mathrm{D}\uparrow)_2$ is not an observable, and is given here for diagnostic purposes only. For tritium, we predict a positive singlet scattering length, $a_s = 34.6$ a.u. (35.8 a.u.), and a large negative s-wave scattering length, $a_t = -82.1$ a.u. (-81.9 a.u.). The T↑ dimer does not possess a bound state, but this large negative a_t indicates that it is only "slightly short of binding."

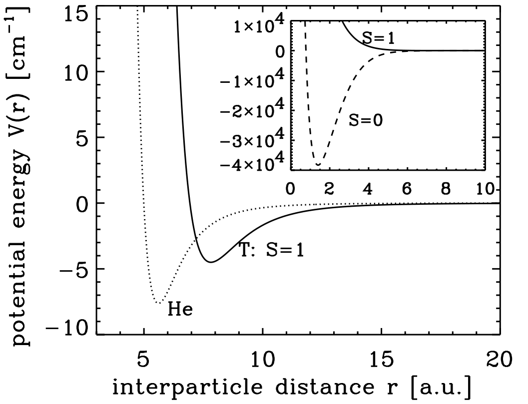

FIG. 1. Tritium dimer triplet $b^3\Sigma_u^+$ potential (solid line) together with the He dimer potential (dotted line) as a function of the interparticle distance r. The inset compares the tritium dimer triplet potential ($S = 1$, solid line) with the tritium dimer singlet ground state potential ($S = 0$, dashed line). Note the different vertical scales of the main figure and the inset. See text for a detailed discussion.

Despite the fact that the tritium triplet scattering length is negative — implying an unstable condensate — it may be possible to form a stable tritium condensate utilizing a Feshbach resonance. Coupled-channel scattering calculations that couple the singlet and triplet subspaces reveal such a Feshbach resonance, i.e., a diverging scattering length for two atoms characterized by quantum numbers F and m_F, as a function of the magnetic field strength. The coupling arises through the atomic hyperfine interaction, and has to be accounted for by an effective two-atom Hamiltonian [16]. Feshbach resonances have been observed experimentally for ^{23}Na and ^{85}Rb [5] among others.

We find that collisions between two tritium atoms, each in their $|F, m_F\rangle = |0, 0\rangle$ state, result in a scattering length of $a(00, 00) = 57$ a.u. (64 a.u.) for zero magnetic field strength B, using the atomic (nuclear) mass. As B increases, the scattering length $a(00 + 00)$ rises and eventually goes through infinity across a broad Feshbach resonance centered at $B = 870$ G (810 G) (see Fig. 2). We also looked for, but could not find, a similar resonance for collisions of hydrogen atoms. Note that our predictions are not sensitive to a replacement of the atomic mass by the nuclear mass.

Formation of a tritium condensate in the high-field-seeking $|0, 0\rangle$ state would require some variety of nonmagnetic trap, such as the dipole CO_2 laser trap that has already produced a ^{87}Rb condensate [7]. Since the electric dipole polarizability of tritium is only 4.56 a.u., the 12 W CO_2 laser setup of [7] would only produce trap depths of the order of a few microkelvin. There appears to be no reason why much stronger CO_2 lasers could not be utilized, however. Realistically, the formation of a tritium condensate will probably require laser intensities at least an order of magnitude more intense. The large magnitude of the zero-field scattering length would make evaporative cooling far more effective than is the case for spin-polarized hydrogen. Another possible way to cool the spinless substate of tritium would be to implement a recent proposal to cool an atomic gas through magnetic field ramps across a Feshbach resonance [17]. Despite these technical difficulties to be overcome, recent improvements in trapping and cooling technology would appear to make the creation of a tritium condensate a viable possibility.

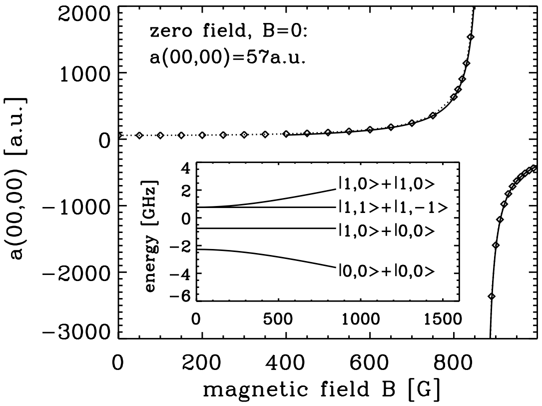

FIG. 2. $a(00+00)$ scattering length (diamonds; using the reduced atomic mass in the coupled-channel calculation, see text) as a function of the magnetic field strength B (dotted lines are shown to guide the eye). The solid line describes the behavior for the range $B \in [400, 1300\ \mathrm{G}]$ well using the following parametrization, $a(00,00) = a_{\mathrm{BG}}[1 - \Delta/(B - B_R)]$ with $a_{\mathrm{BG}} = -37.9$ a.u., $\Delta = -1238$ G, and $B_R = 870.8$ G; however, the fit is inaccurate at low fields. Inset: Threshold energies in GHz as a function of magnetic field B in Gauss. The assignment of quantum numbers is approximate, except for $|1,1\rangle + |1,-1\rangle$, which is an exact eigenstate without admixtures.

To determine the bound state properties of $(\mathrm{T}\uparrow)_N$ clusters with $N > 2$, we first have to investigate the importance of nonadditive contributions to the many-body interaction potential. Although three-body contributions, i.e., the Axilrod-Teller term [18] and three-body exchange terms [19], are significant for the hydrogen trimer in its electronic ground state, they should be less important for the spin-polarized trimer since the classical atom-atom equilibrium distance of the quartet trimer is more than 5 times as large as that for the doublet ground state trimer. Our calculations show that inclusion of the damped Axilrod-Teller term [20] raises the ground state energy of the spin-polarized tritium trimer by roughly 1.6%, and that of the larger clusters slightly more, e.g., by about 6% for $N = 40$. To describe $(\mathrm{T}\uparrow)_N$ clusters, we assume in the following a simple pairwise additive potential energy surface, $V = \sum_{i<j}^{N} V(r_{ij})$, where $V(r)$ denotes the triplet $b^3\Sigma_u^+$ two-body potential. Conceivably, a more sophisticated many-body potential energy surface, which includes effects beyond the two-body potential, could modify our quantitative results somewhat, but we do not expect qualitative changes.

For the $\mathrm{T}\uparrow$ trimer, we use the adiabatic hyperspherical representation [21]. Including only one adiabatic channel yields a single bound state with energy $-1.60 \times 10^{-3}\ \mathrm{cm}^{-1}$. Coupling 25 adiabatic channels results in an energy of $-3.19 \times 10^{-3}\ \mathrm{cm}^{-1}$ with an uncertainty of $10^{-5}\ \mathrm{cm}^{-1}$ and still no excited states. In the limit that an infinite number of channels are coupled, the bound state energy becomes exact, so the uncertainty is the result of including a finite number of channels. There is thus a single $L = 0$ bound state for quartet tritium (L is the total orbital angular momentum); no $L > 0$ bound states are expected since none exist for the ^{4}He trimer [22].

Since the $\mathrm{T}\uparrow$ dimer is unbound, the $\mathrm{T}\uparrow$ trimer is a Borromean state [23]. One may then ask: Does the $\mathrm{T}\uparrow$ trimer state have Efimov character [24]? To investigate this question, we apply a simple quantitative criterion [25], although others exist [26]. If the bound state disappears when one makes the potential more attractive (here achieved by simply multiplying the two-body potential with an overall scaling factor greater than 1), then the state under investigation is an Efimov state; if the bound state does not disappear, it is not an Efimov state. Applying this criterion, our coupled-channel calculations indicate that the bound $\mathrm{T}\uparrow$ trimer state is not an Efimov state. In short, the $\mathrm{T}\uparrow$ trimer has exactly one $L = 0$ bound state, a Borromean state that is highly diffuse spatially.

To calculate the energetics and structural properties of $(\mathrm{T}\uparrow)_N$ clusters with up to $N = 40$ atoms, we employ the diffusion quantum Monte Carlo (DMC) technique [27]. This method solves the time-independent many-body Schrödinger equation essentially exactly, to within a statistical error. Here, we employ the DMC method with importance sampling [27], using a descendant weighting scheme [28] for the extrapolation of structural properties. Our guiding wave functions [27], which enter the DMC calculation, have the analytical form given in Eq. (5) of Ref. [29], and recover between 84% and 96% of the DMC ground state energy when used in a variational quantum Monte Carlo calculation.

For comparison, we find a DMC binding energy of $-2.9(5) \times 10^{-3}\ \mathrm{cm}^{-1}$ for the $\mathrm{T}\uparrow$ trimer, in agreement with our hyperspherical calculation. The number in brackets denotes the statistical uncertainty. Figure 3 compares the ground state energy per particle E_0/N of $(\mathrm{T}\uparrow)_N$ clusters (plusses) with those of $^4\mathrm{He}_N$ clusters (using the LM2M2 potential [14], diamonds). This shows that $(\mathrm{T}\uparrow)_N$ clusters are even more weakly bound than $^4\mathrm{He}_N$ clusters with the same number of atoms.

The comparison between $(\mathrm{T}\uparrow)_N$ and $^4\mathrm{He}_N$ clusters can be extended by considering their structural properties. For example, we find that the $(\mathrm{T}\uparrow)_5$ cluster has an average interparticle distance of $\langle r_{ij} \rangle$ of 22.2 a.u.; the $^4\mathrm{He}_5$ cluster, on the other hand, is $\langle r_{ij} \rangle = 13.6$ a.u. Even if one takes into account that the classical equilibrium distance of the tritium triplet potential is about 2.2 a.u. larger than that of the He dimer potential, the difference between the expectation values of the interparticle distance for these $N = 5$ clusters indicates that the spin-polarized tritium system is even more diffuse than the $^4\mathrm{He}_5$ cluster. To illustrate this aspect further, the inset of Fig. 3 compares the pair distribution of the $(\mathrm{T}\uparrow)_5$ cluster (solid line) with that of the $^4\mathrm{He}_5$ cluster (dotted line). Clearly, the pair distribution of the $(\mathrm{T}\uparrow)_5$ cluster is much broader than that of the $^4\mathrm{He}_5$ cluster. We find similar behavior for clusters with more particles.

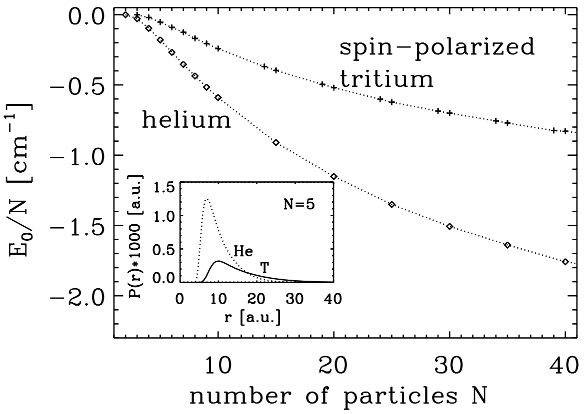

FIG. 3. DMC ground state energy per particle E_0/N for $(\mathrm{T}\uparrow)_N$ clusters (plusses) and $^4\mathrm{He}_N$ clusters (diamonds) as a function of N. Dotted lines are shown to guide the eye. Inset: Pair distribution $P(r)$ for $(\mathrm{T}\uparrow)_5$ cluster (solid line) and $^4\mathrm{He}_5$ cluster (dotted line), calculated by the DMC technique with importance sampling. $P(r)$ is normalized such that $\int_0^\infty P(r) r^2\, dr = 1$. This figure illustrates that $(\mathrm{T}\uparrow)_N$ clusters are even more weakly bound than He_N clusters.

The tritium trimer is the smallest spin-polarized cluster (the dimer is unbound). As discussed above, the two-body potential for spin-polarized hydrogen is almost identical to that of tritium. The hydrogen atom, however, is about a factor of 3 lighter. Consequently, there is no bound state for the H $\uparrow$ trimer. An interesting question to ask is the following: How many atoms are needed to form a bound system of spin-polarized hydrogen atoms? Initial exploratory studies show that more than 100 atoms are needed to form a bound spin-polarized hydrogen cluster. The smallest spin-polarized hydrogen cluster could then be thought of as a "super-Borromean" cluster for which all smaller subsystems are unbound. A detailed study will be published elsewhere.

In summary, this Letter proposes a number of intriguing possibilities for the physics of tritium systems. We point out the possibility for forming a tritium condensate with controllable parameters via a Feshbach resonance. Down the road, one can imagine trapping an atomic hydrogen gas together with an atomic tritium gas, or possibly including deuterium to study fermion systems. We further found that the spin-polarized trimer possesses a Borromean, or halo, state. In addition, we mapped out the properties of larger "exotic" spin-polarized tritium clusters. Studies of spin-polarized clusters are interesting by themselves [30], as they enter new many-body physics regimes. For instance, unexpected physics may emerge from manipulating the two-body interaction via the Feshbach resonance in a cluster. In short, tritium offers a wealth of interesting physics by virtue of its weak attraction.

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