Vth Workshop on

QUANTUM FLUID CLUSTERS

ECT*, Trento (Italy)

September 16 – 21, 2002

Book of abstracts

M. Barranco, F. Dalfovo and J. Navarro, Editors

PROGRAM

Monday 16

9:00-9:40 Registration
9:40-9:50 M. Barranco: Opening
9:50-10:30 K. von Haeften: *Bubble formation and energy relaxation in electronically excited ^{3}He- and ^{4}He-clusters* 40

COFFEE BREAK

11:00-11.40 H. Maris/M. Pi: *Optical Properties of Electron Bubbles in Liquid Helium* 31/34
11:40-12:20 M. Lewerenz: *Quantum Monte Carlo Simulations with on the Fly Potentials: Structural Quantum Effects in $(NeHe_n)^+$* 28

LUNCH BREAK

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BREAK

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16:50-17.30 C.P. Schulz: *Dynamics of Rb and Cs doped helium nanodroplets* 11

Tuesday 17

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9:50-10:30 R. Schmied: *Quantum Rotation in Helium Clusters: a PIMC Study* 37

COFFEE BREAK

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11:40-12:20 F.A. Gianturco: *Flying refrigerators: atomic and molecular impurities in small ^{4}He droplets* 14

LUNCH BREAK

15:00-15:40 F. Stienkemeier: *Alkali Clusters in Giant Spin States from Helium Nanodroplet Isolation* 41

BREAK

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16:50-17:30 K. Lehmann: *The Quantum Hydrodynamic Model for the Motion of Solutes in Liquid Helium and Angular Momentum Conserving Evaporative Cooling of Helium Nanodroplets* 27

Wednesday 18

9:10-9:50 Y. Kwon: *Structure and spectroscopy of OCS in Helium, Hydrogen and mixed Helium/Hydrogen clusters* 43
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COFFEE BREAK

LUNCH BREAK

Thursday 19

COFFEE BREAK

LUNCH BREAK

BREAK

Friday 20

COFFEE BREAK

LUNCH BREAK

BREAK

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9:50-10:30	von Haeften	Schmied	Dalfovo	Gordillo	Krotscheck
10:30-11:00	COFFEE BREAK				
11:00-11:40	Maris/Pi	Wyatt	Galli	Reatto	Poves
11:40-12:20	Lewerenz	Gianturco	Boronat	Leadbeater	Stringari
12:20-15:00	LUNCH BREAK				
15:00-15:40	Garcias	Stienkemeier	Sarsa	Tiggesbäumker	Zillich
15:40-16:10	BREAK				
16:10-16:50	Ernst	Miller		Ancilotto	Vilesov
16:50-17:30	Schulz	Lehmann		Draeger	Toennies

Each talk has alloted 30 min plus 10 min for discussion

Social event
A Conference dinner will be offered at the restaurant (Rifugio) Bindesi on Thursday.

Financial support

European Centre for Theoretical Studies in Nuclear Physics and Related Areas (ECT*).

Dipartimento di Matematica e Fisica, Università Cattolica del Sacro Cuore, sede di Brescia, and Dipartimento di Fisica, Università di Trento, through fundings of MIUR-PRIN2000 "Theory of coherent fluids: Bose gases, superfluids and superconductors".

ABSTRACTS

Dynamics of Rb and Cs doped helium nanodroplets

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Helium nanodroplets (He_N, $N \approx 5000$) are doped with alkali metal atoms which reside weakly bound to the surface of the droplets. We apply time-resolved femtosecond pump-probe spectroscopy in order to study the dynamics of the excited alkali atom interacting with the helium surface. Here we present our recent experiments on the formation of exciplexes in Rubidium and Cesium doped droplets, which is an extension of our previous work on potassium doped helium droplets [1].

In the experiment the first (pump) laser pulse is used to electronically excite the alkali atom on the helium droplet's surface. After a variable delay time in the range of femto- to picoseconds the second (probe) laser pulse photo-ionizes the excited atoms and possible excited reactions products. The ions are mass-selected in a quadrupole analyzer. For Rb-doped droplets we found that Rb*He exciplexes are only formed when exciting the ${}^2P_{3/2}$ state. No exciplexes have been observed from the excited ${}^2P_{1/2}$ state in agreement with recent laser-induced fluorescence studies [2]. We measured a formation time of Rb*He in the ${}^2P_{3/2}$ state of 44 ps. This is significantly longer when compared to 170 fs, the formation time of K*He exciplexes, found in our previous experiments [1]. Hence, our results confirm the prediction of Reho et al. [3]. When repeating the experiment with cesium doped nanodroplets no exciplexes have been observed again in agreement with the prediction of Reho et al.

[1] C.P. Schulz, P. Claas ans F. Stienkemeier, Phys. Rev. Lett. **87**, 15340 (2001).

[2] F.R. Brühl, R.A. Trasca and W.E. Ernst, J. Chem. Phys. **115**, 10220 (2001).

[3] J. Reho, J. Higgins, C. Callegari, K. Lehmann, and G. Scoles, J. Chem. Phys. **113**, 9686 and 9694 (2000).

Quantized vortices in Helium droplets?

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We would like to discuss the appearance and possibility of creating quantized vortices in superfluid helium drops [1, 2], and to compare the situation with that of Bose-Einstein condensates of alkali gases.

In the case of helium drops, within density functional (DF) theory we will address the structure of pure ^{4}He and mixed ^{3}He-^{4}He droplets in which an impurity, typically an inert gas atom or a linear molecule like HCN, has been embedded [3, 4].

The role played by the impurity in the pinning of the vortex line will be discussed, as well as the robustness of the dopant+vortex+$^4\mathrm{He}_{N_4}$ complex when ^{3}He is added [3, 4]. The binding energy of the dopant to the vortex line will also be discussed.

[1] G. H. Bauer et al., J. Low Temp. Phys. **98**, 47 (1995).

[2] J. D. Close et al., J. Low Temp. Phys. **111**, 661 (1998).

[3] F. Dalfovo et al., Phys. Rev. Lett. **85**, 1028 (2000).

[4] R. Mayol et al., Phys. Rev. Lett. **87**, 145301 (2001).

Femtosecond dynamics of metal clusters embedded in helium droplets exposed to intense laser beams

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A beam of silver cluster doped helium droplets is produced by means of a supersonic expansion of pure helium gas through a small nozzle [1]. The droplet beam passes a vapor cell and collects silver atoms. In this manner silver clusters consisting of up to about 130 atoms can be produced. Additional doping with rare gas atoms can be achieved by an inlet in the scattering chamber. The measurement have been performed with the resonant two photon ionization technique (R2PI) using a tunable laser system. At an photon energy of 3.97 eV Ag_8 is resonantly ionized [2] and the properties of this transition state can be investigated.

In a first set of experiments we studied the influence of an rare gas nanomatrix (RG) around this cluster ($Ag_8RG_N@He_{droplet}$) [3]. In contrast to the red–shiftobtained for argon, krypton and xenon, for neon a net shift of the resonance to shorter wavelengths is observed. It was found that the shift of the resonance energy is sensitive to the size, the dielectric constant, and the atomic electron affinity of the nanomatrix. This behavior can be explained by a simple qualitative model [3].

In metal nanoparticles the absorption is dominated by the collective excitation of the valence electrons. The lifetime was found to be in the femtosecond regime. In order to determine whether the transition state in Ag_8 is molecular– or more plasmon–like we have performed pump–probe measurements using the resonant two–color ionization technique [4]. It turns out that delayed nanosecond light pulses are sufficient to achieve a reasonable time resolution and the lifetime of the resonance at 312 nm was found to be about $4\mu s$.

Resonant two photon ionization was also used to obtain the first photoelectron spectra of size–selected clusters inside helium droplets [5]. From the spectra the ionization potential of $Ag_8@He_{droplet}$ can be extracted and a value of 6.7 eV was found. In addition, the electron signal shows a dependence on the droplet size. This indicates a strong influence of the helium droplet environment on the electron emission process.

[1] A. Bartelt, J.D. Close, F. Federmann, N. Quaas, and J.P. Toennies, Phys. Rev. Lett. **77**, 3525 (1996).

[2] F. Federmann, K. Hoffmann, N. Quaas, and J.P. Toennies, Eur. Phys. J. D **9**, 11 (1999).

[3] Th. Diederich, J. Tiggesbäumker and K.-H. Meiwes–Broer, J. Chem. Phys. **116**, 3263 (2002).

[4] Th. Diederich, T. Döppner, J. Tiggesbäumker, and K.-H. Meiwes–Broer, in preparation.

[5] P. Radcliffe, Th. Diederich, T. Döppner, A. Przystawik, J. Tiggesbäumker, and K.-H. Meiwes–Broer, in preparation.

Flying refrigerators: atomic and molecular impurities in small ^{4}He droplets

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To analyse the structural behaviour of small atomic and molecular impurities in helium droplets requires both a reliable knowledge of the interaction forces and a quantum treatment of the formation of the many-particle bound states as a function of the droplet size, taking advantage of the special behaviour of the quantum solvent represented by the bosonic adatoms of ^{4}He.

In our research group we have attempted, in the last few years, to work on both fronts by developping a parameter-free modelling of the weak van der Waals (vdW) forces between simple molecular dopants like OCS with He and H_2, and CO with both ^{4}He and Ar [1, 2, 3, 4]. For such systems we have further tried to test as much as possible the quality of the final potential energy surfaces (PES) by using them to generate inelastic cross sections [4], to evaluate transport properties of the systems [5] and to compare the computed rovibrational structures of the one-atom complexes with the available experiments [6].

The next step has been to take advantage of stochastic methods, in particular of Diffusion Montecarlo (DMC) methods, to generate the ground state configurations of the bound systems by adopting the above PES, plus the atom-atom interactions for the adatoms, or by choosing existing atom-dopant interaction potentials for impurities like Li, H^-, Na and H [7]. By constructing the many-particle potentials as sums of dopant-He interactions plus He-He interactions, we have therefore analysed the spatial structures, the binding energies and the solvent effects on molecular properties (e.g. vibrational and rotational shifts) as a function of the cluster size and of the competing effects of the various potentials in controlling the "solvation" of the dopant particle within the ^{4}He droplets [8, 9].

The results on both the energetics and the structural behaviour as a function of both dopant system and droplet size will be presented at the meeting, together with a discussion of the modelling procedure employed to obtain our DFT-DISP potential energy surfaces.

[1] F.A. Gianturco and F. Paesani, Mol. Phys. **99**, 689 (2001).

[2] F.A. Gianturco and F. Paesani, J. Chem. Phys. **113**, 3011 (2000).

[3] F.A. Gianturco and F. Paesani, M. Lewerenz, and J.P. Toennies, J. Chem. Phys. **111**, 9897 (1999).

[4] E. Bodo, F.A. Gianturco and F. Paesani, Zeit. Phys. Chem. **214**, 1013 (2000).

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[7] C. Di Paola, F. Paesani and F.A. Gianturco, J. Phys. B, submitted (2002).

[8] F. Paesani, F.A. Gianturco and K.B. Whaley, J. Chem. Phys. **115**, 10225 (2001).

[9] F. Paesani and F.A. Gianturco, J. Chem. Phys., submitted (2002).

Superfluidity in a Doped Helium Droplet

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Path Integral Monte Carlo calculations of the superfluid density throughout ^{4}He droplets doped with linear impurities $(HCN)_n$ are presented. After deriving a local estimator for the superfluid density distribution, we find a decreased superfluid response in the first solvation layer. This effective normal fluid exhibits temperature dependence similar to that of a two-dimensional helium system. The relationship between the induced normal fluid and the effective moment of inertia is discussed.

Microscopic Dynamics in Bulk Superfluid Helium: Experiment and Theory

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To investigate implications of superfluidity on microscopic scales, we carry out time resolved studies of molecular dynamics in bulk superfluid helium. To this end, we have devised several experimental schemes for the injection of molecular impurities in bulk superfluid helium. Among these, the combination of a cryogenic pulsed valve with laser ablation has been most extensively developed as a universal molecule injector. Progress in these developments will be described. However, todate, the most complete experiments remain time-resolved studies of the intrinsic excitations in neat liquid helium, namely the excimeric Rydberg states. These molecular centers serve as nanoscale transducers to drive the liquid into motion, and as nanoscale viscosimeters that allow the measurement of the time resolved liquid response through fluorescence depletion spectroscopy. The observed real-time dynamics can be understood in terms of the two-fluid hydrodynamics, with the conclusion that these phenomenological equations of motion scale down to the molecular size scale. To provide the quantum framework for the observed dynamics, and to address subtleties of interactions and the validity of approximations used to implement classical hydrodynamics, we have developed time-dependent density functional simulations of the bulk liquid. We have applied this treatment to the classic problem of the relaxation dynamics of an excess electron in superfluid helium. The careful mapping between classical two-fluid dynamics and quantum dynamics of the well defined system validates the approximations made in our prior analysis. The dynamics of relaxation of the liquid cavity around the excess electron, i.e. the formation of the electron-bubble, will be discussed. We find this process to occur on a significantly longer time scale than what has been believed previously.

Alkali-ion-doped clusters of ^{4}He

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A positive alkali ion in liquid ^{4}He polarizes the neutral Helium atoms, that are attracted towards the impurity where the electric field is stronger [1]. This phenomenon by which the atoms of the liquid coalesce around the impurity is at the basis of the reduced mobility of such ions in the bulk liquid. This is relevant for instance to the experiments that probe Landau's critical velocity in the superfluid by the drift velocities of charged species with an applied electric field [2]. The effective microscopic structure of a doped cluster is therefore the result of the delicate balance between competing interactions: condensation due to the strong attractive part of the ion-Helium potential and hard-core repulsion, both between the ion and the surrounding atoms and between the Helium atoms themselves. We will report on our Variational and Path Integral Monte Carlo simulations of an alkali ion impurity (Na^+, K^+ and Cs^+) in liquid ^{4}He droplets. We will give results on the microscopic structure of the doped cluster in terms of the radial density profile around the ion and of angular correlations. In all cases, we find that the positive ion resides stably at the center of the droplet and modulates the Helium density. The most distinctive feature is the presence of a well marked inner shell of Helium atoms close to the ion, whose location and width depends on the chemical species of the core ion. The radial density profiles and angular correlations show that the microscopic structure of the snowball is remarkably different for each ion. We are working also on simulations of two alkali ions in a ^{4}He droplet.

[1] K.R. Atkins, Phys. Rev. **116**, 1339 (1959).

[2] P.V.E. McClintock, Physica **127B**, 300 (1984).

Structure and elementary excitations of doped ^{3}He clusters

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To understand how various bulk physical properties of quantum liquids are modified in restricted geometries, helium drops have been the object of extensive experimental and theoretical investigations [1]. In this work, ground state properties and multipole response of $^3\mathrm{He}_N$ drops doped with inert gas atoms and SF_6 molecules have been investigated in the framework of the Finite-Range Density Functional Theory and the Random Phase Approximation.

We have found that the solvation energies are smaller than in the case of $^4\mathrm{He}_N$ doped drops, but still negatives. Due to the attraction exerted by the impurity on the surrounding ^{3}He atoms, doped drops of any size are bound [2].

Concerning the multipole collective excitations, the presence of an attractive impurity increases the spectrum fragmentation and decreases the mean energy of surface modes, as compared with the results obtained for pure $^3\mathrm{He}_N$ drops. For large clusters the mean energy of surface modes lies below the atom emission threshold, whereas for the monopole volume mode it is always above the threshold. When the cluster size increases the dipole mean excitation tends to zero, indicating that the impurity is delocalized in the bulk of the drop [3, 4].

[1] J.P. Toennies and A. Vilesov, Annu. Rev. Phys. Chem. **49**, 1 (1998), and references therein.

[2] F. Garcias, Ll. Serra, M. Casas, and M. Barranco, J. Chem. Phys. **108**, 9102 (1998).

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[4] F. Garcias, Ll. Serra, M. Casas, and M. Barranco, J. Chem. Phys. **115**, 10154 (2001).

Time-dependent Density Functional calculations of dynamical properties of liquid ^{4}He in confined geometries

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Atoms and ions immersed in superfluid ^{4}He are successfully used as microscopic probes to investigate the fascinating properties of this quantum system [1]. Most atomic impurities trapped in a ^{4}He cluster have their stable state in the center of the cluster itself, where they are completely surrounded by He atoms. A notable exception to this behavior is represented by alkali metal impurities, which reside in a "dimple" on the surface of the cluster, rather than being solvated inside [2].

Another interesting aspect of liquid ^{4}He is its behavior when adsorbed on a solid surface. Although most of the solid surfaces are wet by He already at $T \sim 0$, on very weakly-attractive surfaces (like those of heavy alkali metals) the most stable state is represented by macroscopic droplets on an otherwise dry surface [3]. As the temperature increases the droplet may spread to cover the whole surface ("wetting" transition).

In recent years Density Functional (DF) methods have become increasingly popular among the theoretical tools available for the study of structural and thermodynamic properties of classical and quantum fluids because of their ability to describe inhomogeneous systems and phase equilibria also in the presence of external fields. This is the case, for instance, of a ^{4}He cluster doped with atomic/molecular impurities or of a ^{4}He film/droplet adsorbed on a solid surface.

We have extended our previous DF calculations for the *static*, equilibrium properties of the ^{4}He systems described above, based on a phenomenological Density Functional proposed in Ref. [4], to the more challenging domain of real time evolution. We have studied in particular, by means of numerical simulations within the Time-Dependent Density Functional Theory, the oscillations of impurities trapped in "dimple" states on the surface of ^{4}He clusters, and the dynamics of ^{4}He droplets/films adsorbed on "weak" surfaces.

Our calculations are fully three-dimensional, allowing in principle for arbitrary deformations of the liquid He structure like those occurring in the above systems.

[1] J.P. Toennies, A.F.Vilesov, Annu. Rev. Phys. Chem. **49**, 1 (1998).

[2] F. Stienkemeier, J. Higgins, W.E. Ernst, and G. Scoles, Phys. Rev. Lett. **74**, 3592 (1995); F.Ancilotto, E.Cheng, M.W.Cole, and F.Toigo, Z. Phys. B **98**, 323 (1995).

[3] E.Cheng, M.W.Cole, W.F.Saam, and J.Treiner, Phys. Rev. B **46**, 13967 (1992).

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Two-dimensional H_2 puddles: A PIMC study

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The possibility of maintaining H_2 in a fluid state at temperatures low enough to produce a superfluid has recently attracted considerable attention. Unfortunately, the H_2-H_2 interaction is too strong to allow molecular hydrogen to be a liquid below its bulk triple point of 14 K.

One way to attain a liquid ground state is to reduce the effective attraction between the H_2 molecule by reducing the number of neighbors. That can be done by considering a 2D or a quasi one dimensional environment [1, 2]. A second way is to consider 3D clusters [3], or finaly, to create 2D puddles on top of an adequate substrate [4]. We study here strictly 2D H_2 clusters with sizes ranging from $N = 6$ to $N = 61$ and determine the fraction of molecules considered to be superfluid (*i.e.* decoupled from motions of the substrate) as a function of temperature. We checked also the relation between superfluidity and cluster structure. This was found to be layered, with molecules distributed in shells around the center of mass of the clusters. The maximun values of the superfluid density are obtained in clusters with less than two concentric layers. There is also a decrease in the mobility of the molecules in going from two-layered clusters to three-layered ones, what suggests the existence of a solid-like inner core together with a like-like external one.

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[2] M.C. Gordillo, J. Boronat and J. Casulleras, Phys. Rev. Lett. **85**, 2348 (2000).

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New Phenomena in Expansions of Solid and Liquid Helium into Vacuum

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In a molecular beam apparatus we have studied the particle beams produced in expanding helium through 2 micron diameter orifices into a vacuum system. The angular and velocity distributions as well as the total flux have been measured for source pressures P_0 between 0.2-80 bar and source temperatures T_0 between 1.5 and 4.2 K. With liquid in the source at $P_0 < 25$ bar the beam has a narrow velocity distribution with a relative half-width of less than 1%. On lowering the temperature the angular half-width decreases and at 1.5 K is less than $2 \cdot 10^{-3}$ rad. Microscope pictures indicate that the beam consists of large droplets of diameters greater than about 0.5 microns. By increasing the nozzle diameter to 10 microns a continuous coherent quantum liquid jet of several mm length can be expected. Thus this new source opens up many exciting possibilities for a variety of new experiments.

At higher pressures the helium in the source solidifies. Nevertheless a beam of atoms and clusters consisting of a continuous and an intermittent component with a period between 50 and 600 sec depending on the pressure is observed. The theory for this new "geyser effect" predicts that the period and temporal shape of the pressure bursts provide new insight into vacancy mobilities and diffusion and recombination coefficients. Experiments with non-superfluid ^{3}He are in progress to explore the possibility that this new effect may be related to observe the long sought-after "supersolid" phenomenon.

On the stability of small mixed clusters of ^{4}He and ^{3}He

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The stability of small helium clusters results from a delicate balance between the large zero point motion and the extremely weak interatomic potential. Thus the binding energy of the dimer ^{4}He$_2$ is only of about 1 mK [1]. In contrast, the minimum number of atoms of ^{3}He required to form a self-bound system has been theoretically estimated to be around 30 atoms [2, 3, 4], as a consequence of the smaller atomic mass and the Pauli principle.

In this contribution we analyze the existence of small clusters made of few ^{4}He atoms with an increasing number of ^{3}He atoms. The main findings are: i) There exists a "stability island" of clusters with two ^{4}He atoms and up to three ^{3}He atoms, but the system ^{4}He$_2$–^{3}He$_3$ is metastable; moreover the cluster ^{4}He$_2$–^{3}He$_{20}$ is bound. ii) All drops obtained by adding ^{3}He atoms to the trimer ^{4}He$_3$ are bound, but the clusters ^{4}He$_3$-^{3}He$_{3,4,5}$ are again metastable. iii) all remaining clusters with four or more bosons and any number of fermions are stable.

These results are based on a variational calculation using trial wave functions which includes short-range Jastrow correlations, different for boson-boson, fermion-fermion and boson-fermion pairs. Moreover, medium and long range pair correlations are taken into account within a Configuration-Interaction scheme.

[1] R.E. Grisenti, W. Schöllkopf, J.P. Toennies, G.C. Hegerfeldt, T. Köhler, and M. Stoll, Phys. Rev. Lett. **85**, 2284 (2000).

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[4] R. Guardiola, Phys. Rev. **B62**, 3416 (2000).

Curvature and shape effects on adsorbed helium mixtures

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Departures of helium fluids and mixtures from spatial homogeneity take place in many different fashions, among which, droplets and curved twodimensional manifolds adsorbed in nanotubes and other cylindrical pores, present many aspects which can be traced to curvature effects. In particular, clusters of helium mixtures deposited on adsorbing substrates offer an interesting illustration of the competition between selfsaturation, mutual interaction and external confinement. Our density functional calculations show that small amounts of ^{3}He atoms, added to an axially symmetric ^{4}He drop deposited on a flat cesium surface at zero temperature, populate bound states localized at the contact line. These edge states are structurally different from the well-known Andreev states which appear at the top and at the liquid-solid interface of films, since they originate in the peculiarities of the single-particle mean field for this specific geometry, governed by the finite number of particles, their interactions and by the adsorbing potential. We are able to compute the one-body density of up to $N_3 = 200$ ^{3}He particles on a drop with 1000 ^{4}He atoms, and observe that for sufficiently large N_3, the density profiles spread beyond the edge, coating both the curved drop surface and its flat base, eventually isolating it from the substrate. In this context, one can revisit the main features of wetting of weak alkali adsorbers by the helium isotopes at zero temperature, and examine the crossover between the nonwetting behavior of ^{4}He and the wetting abilities of ^{3}He.

Dynamics of Rubidium and Cesium Atoms on Helium Nanodroplets

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Alkali atoms are deposited on superfluid helium droplets where they stay at the surface and form molecules in cold collisions [1]. Through evaporation of helium atoms from the droplet, the temperature is automatically maintained at 0.4 K. Any spectroscopic measurement in the helium beam downstream (with respect to the region of atomic pick-up and reaction) is particularly sensitive to weakly bound species since the formation of molecules with large bond energy leads to a strong helium depletion and disappearance of the droplet from the beam. In alkali doped droplet beams, alkali dimers are preferentially detected in triplet states and trimers in quartet states.

Our experiments with rubidium and cesium allowed the observation of Rb_2 and Cs_2 triplet transitions as well as RbHe exciplex spectra. Emission spectra reveal the presence of vibrational relaxation and nonadiabatic intersystem crossings that result from the proximity of the helium droplet surface. Cesium atoms were excited to the $7\ ^2P$ state in order to initiate a reaction with H_2 molecules on the droplet. The current state of our investigations will be reported and its potential use for the study of collisional processes in other trapping experiments discussed.

[1] See special issue of the Journal of Chemical Physics, Vol. 115 (2001), on *Helium Nanodroplets: A Novel Medium for Chemistry and Physics.*

Elastic and inelastic processes in droplets, films, and slabs

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We discuss, on a microscopic level, the effects of confinement on structural as well as dynamic properties of quantum liquids. The most evident *structural* consequences of confinement are *layer structures* found in liquid films, and *free surfaces* appearing in liquid drops and slabs. These structural properties have immediate consequences: new types of excitations such as surface phonons, layer phonons, layer rotons and standing waves can appear and are potentially observable in neutron scattering spectra as well as in thermodynamic properties.

Atom scattering experiments provide further insights into structural properties. Methods have been developed to describe elastic and inelastic atom scattering as well as transport currents. The theory has been applied to examine scattering processes of ^{4}He and ^{3}He atoms impinging on ^{4}He clusters, as well as ^{4}He scattering off ^{4}He films and slabs. In all cases, we find that *inelastic* processes dominate scattering events.

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Vortex creation and interaction in uniform and trapped condensates.

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We will present results for vortex creation by the motion of massive objects in Bose-Einstein condensates, modelled by the Gross-Pitaevskii equation. We will first present results for vortex creation in uniform systems and show how the system evolves through a sequence of stationary states [1]. By including background density fluctuations we demonstrate how the critical velocity for vortex formation is reduced with decreasing ground state population. Finally we show how this situation is modified for the case of a non-uniform condensate. The possible relaxation mechanisms of the produced vortex system are also discussed.

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The Quantum Hydrodynamic Model for the Motion of Solutes in Liquid Helium and Angular Momentum Conserving Evaporative Cooling of Helium Nanodroplets

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At the previous conference in this series, the author presented the results for the calculation of rotational moments of inertia of molecules in liquid helium based upon the kinetic energy induced in an inhomogeneous, ideal fluid by the motion of the helium solvation density as the molecule rotates. This model has since been extended in several ways: (1) The model has been shown to follow from a variational optimization of a trial wavefunction of a Feynman form of the many body ground state times a product of single body phase functions; (2)The model has been shown to provide quantitative agreement with VQMC predictions of the effective mass of alkali cations in helium; (3) A rigorous model for the coupling of translation and rotation has been derived for linear molecules in our work and for general rotors by Quist and Elser.

Portner, Vilesov, and Havenith have observed that tetracene molecules picked up by a helium nanodroplet beam have substantial lab frame alignment, which they interpreted as arising from the "memory" of the direction of the initial collisional angular momentum of the pick up process. The author has developed a statistical evaporation model for helium nanodroplets that includes angular momentum conservation. It is found that for initial conditions of energy and total angular momentum similar to those expected for the pick up of a tetracene molecule, the droplets cool to a final temperature of 0.4 K, but that the angular momentum distribution is much broader than that predicted by thermal equilibrium at this temperature and to be highly dependent upon the initial angular momentum. Further, it is found that for initial angular momentum values above 2000 units, the final alignment of droplet angular momentum is only slightly reduced from the assumed initial condition, $\langle P_2 \rangle = -0.5$

Quantum Monte Carlo Simulations with on the Fly Potentials: Structural Quantum Effects in $(\mathrm{NeHe}_n)^+$

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Many detection schemes for neutral particles embedded in helium clusters rely on ionisation methods. This process usually involves significant fragmentation of the original cluster. While the strong quantum effects in neutral pure or mixed helium clusters prevent the occurrence of magic cluster sizes, such specially stable clusters have been observed for several ionic species in drift tube experiments and in the fragmention following the ionisation of large doped helium clusters.

The similarity of the ionisation potentials of neon and helium allows significant charge delocalisation in mixed clusters of the type $(\mathrm{NeHe}_n)^+$, such that the computation of the vibrational ground state structure on simple additive model potentials is impossible. We have combined an efficient diffusion quantum Monte Carlo code with a DIM (diatomics in molecules) approach for the 'on the fly' calculation of the electronic energy at the large number of particle arrangements required to map out the rovibronic ground state.

Vibrational delocalisation is found to play a major role in $(\mathrm{NeHe}_n)^+$, but indications of shell formation and the onset of magic cluster sizes remain. A tightly bound triatomic $[\mathrm{He\text{-}Ne\text{-}He}]^+$ ion core is surrounded by outer shells of essentially van der Waals bound helium atoms. Particle exchange between the ion core and the outer region seems to be inhibited. The quantum ground state structures are found to systematically differ from the global potential minimum and show higher than expected symmetries.

Dimerization of formic acid dimer

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We have observed the IR spectrum of formic acid dimer in the gas phase and in superfluid liquid helium clusters. Formic acid serves as a prototype for double hydrogen bonded clusters. Whereas the gas phase spectrum of formic acid dimer reveals a symmetric structure with a concerted (out of plane) proton transfer on the time scale of nsec, the dimerization proces in liquid helium droplets results in distinct structures: IR spetroscopy of formic acid dimer in liquid helium droplets clearly indicates the formation of a non symmetric polar species with open C-H and O-H bonds. We compare the observed spectrum with the result of DFT calculations for the different structures. The observed spectrum can be attributed to a local minimum in the potential surface which is reached by the specific dimerization proces at ultracold temperatures. The results of this study can be directly compared with the oberservation of an asymmetric structure by formation in an Argon matrix [1].

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Free surface of superfluid ^{4}He and impurity states

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We present a microscopic description of the static properties of the free surface of liquid ^{4}He at zero temperature using the diffusion Monte Carlo (DMC) method. Relying on a slab geometry and the same Aziz potential that we have previously used in calculations of the bulk, the energy of the system as a function of the number of particles has been calculated. The surface tension can be drawn from the number of particles dependence of the energy, the results being in an overall agreement with recent experimental data. Other characteristic properties of the surface like the density profile, and its possible asymmetry, the surface thickness, the kinetic density profiles, and structure functions have been also studied. The calculation of the one-body density matrices as a function of the distance to the surface has allowed the determination of the condensate density profile $\rho_0(z)$. The results obtained show an enhancement of $\rho_0(z)$ near the surface, a feature already observed in variational calculations.

The second part of the work is devoted to the study of energetic and structural properties of light impurities (^{3}He, H, H_2) on the surface. The results of the impurity density profiles illustrate their most probable location. The three cases analyzed show an energetically favorable localization on the surface, the molecular impurity H_2 being the most penetrating. In order to determine their binding energies, we have used the generalized reweighting technique developed in our group and successfully used in DMC calculations of the chemical potentials of impurities in bulk ^{4}He. The impurity chemical potentials obtained are in reasonable agreement with previous theoretical estimations and available experimental data.

Optical Properties of Electron Bubbles in Liquid Helium

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An electron injected into liquid helium forces open a cavity from which essentially all helium atoms are excluded (electron bubble). When the electron is in its ground state the cavity is spherical, and has a radius of approximately 19 Å for zero applied pressure. It is possible to optically excite the electron to higher energy states; the shape of the electron bubble then changes. We will describe the results of calculations of the shape of the bubbles in the different excited states, and as a function of pressure. Experimental studies of the optical properties of the bubbles will be reported, including a novel technique for the detection of the excited state bubbles. Calculations of the properties of the two-electron bubble will also be presented.

The Growth of Metastable Species in Helium Nanodroplets: Characterization by High Resolution Infrared Laser Spectroscopy

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Superfluid helium droplet spectroscopy is providing new insights into a wide array of structural and dynamical processes, including superfluidity, solvent-solute interactions, rotational and vibrational dynamics and the self assembly of novel new cluster structures. Infrared laser spectroscopy has proven to be particularly useful in these studies, due to the fact that the resolution of the associated spectra is often very high. For these studies, helium turns out to be a nearly ideal matrix for isolating molecules. These methods are now being used in our laboratory to study biomolecules, radicals, ions, hydrogen bonded complexes and metal nanoparticles.

The apparatus built at UNC uses a bolometer to measure the laser-induced change in the helium droplet beam energy. A color center laser operating in the range 2800 - 4500 cm-1 is used to vibrationally excite a molecule within the droplet, which is followed by vibrational quenching of the molecule by the helium and the evaporation of several hundred helium atoms. This reduces the energy delivered by the helium droplets to the bolometer, giving rise to the observed signals.

The focus of the present talk will be on the use of helium nanodroplet spectroscopy in determining molecular structures, from metal nanoparticles to novel cluster structures. We show that cluster growth in liquid helium in fundamentally different from that in the gas phase, providing us with access to new regions of the intermolecular potential energy landscape. In the case of metal particles, we are able to bridge the gap between small systems, where the bonding is purely van der Waals, to larger systems, which mimic the behavior seen in chemisorbed systems. Given that these systems are of finite size, they are ideal for comparison with ab initio theory.

Rotational constants of molecules inside He drops

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A fully microscopic calculation of the effective moment of inertia of the OCS molecule inside ^{4}He droplets is presented. A systematic analysis of the reduction of the rotational constant of the system as a function of the number of helium atoms has been performed. A comparision with the experimental values shows that our results not only reproduce the observed trend but also present a good quantitative agreement. The calculations have been done by carrying out quantum monte carlo simulations. A rigid body model was used for the OCS molecule with rotation along the principal axes of inertia and center of mass translation. Accurate variational wave functions with angular dependence have been obtained. These functions have been used as a guide in a Reptation Quantum Monte Carlo calculation of the energy, density distributions and the effective moment of inertia. The moment of inertia has been obtained by analyzing the the propagation in the imaginary time. A recently developed potential energy surface was employed for the helium-OCS interaction while the accurate SAPT potential was used for helium-helium. The pheomenom of the moment of inertia enhancement is discussed in terms of the density profiles.

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Electron bubble explosions in liquid helium

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We have used a Hartree-type electron-helium potential together with a density functional description of liquid ^{4}He to study the explosion of electron bubbles submitted to a tensile strenght. The critical pressure at which bubbles explode has been determined. It has been found that this critical pressure is very close to the pressure at which the liquid becomes globally unstable in the presence of electrons. Using a capillarity model we have estimated that the quantum to thermal crossover temperature is very small, of the order of 4.7 mK. The critical pressure turns out to be very sensitive to the electron-helium potential and to the helium density profile.

On the structure of ^{3}He shell coating ^{4}He droplets

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Mixed droplets of ^{3}He and ^{4}He have been instrumental in the experimental demonstration of the occurrence of superfluidity in ^{4}He drops of a few tens of atoms [1]. The almost inmiscibility of both fluids produce other novel physical situations that we aim to describe in this presentation. When the number of ^{4}He atoms is large compared to the number of ^{3}He atoms, the latter form an spherical shell/film on the surface of the ^{4}He drop. Results using the density functional formalism show that the mean field to which the helium-3 atoms are subjected can be approximated by a one dimensional harmonic oscillator in the radial coordinate, centered at the surface of the helium-4 droplet, perturbed by a centrifugal term. This gives rise to a new and unexpected shell structure of the form

$$1s, 1p, 1d, 1f, 1g, \ldots 2s, 2p, 2d, 2f, \ldots$$

corresponding to the single particle energy formula

$$E^{nl} = \hbar\omega_0(n + 1/2) + \hbar\omega_1 l(l + 1)$$

with $\omega_1 << \omega_0$. The Fermi level lies always in the $n = 1$ region.

The residual two body interaction between the ^{3}He atoms can be computed using the mean field wave functions and the same interaction used in the density functional calculation. It is qualitatively similar to the interaction found in the case of pure ^{3}He droplets [2] –a spin-spin attractive interaction– although much weaker. As the mean field gaps are also much smaller than in the case of a pure ^{3}He drop, the aligned solution may eventually overcome the shell structure producing bubbles of ^{3}He bearing the maximum allowed spin, equal to one half the number of helium-3 atoms.

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Alkali atoms with negative scattering length: bright soliton states

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It is well known that in one dimension soliton states exist for Bose condensed atoms with negative scattering length described by the Gross-Pitaevskii equation. Such states, called bright solitons, are characterized by a time invariant shape. We have studied the existence of similar states in higher dimensions. In two and three dimensions in a flat potential solitons are not stable. However if an external harmonic potential constrains the motion in two dimensions such soliton states are stable but there is a maximum number N_c of bosons otherwise the system collapses. The value of N_c depends on the transverse confinement, more precisely it is proportional to the ratio of the transverse harmonic length to the scattering lenth. Such bright soliton has some similarity with a cluster but its shape is anisotropic due to the transverse confinement. This anisotropy decreases as the number N of bosons approaches the critical value N_c. We have investigated the stability and the collective oscillations of such bright solitons. We have also investigated the reflection and the tunneling of a bright soliton impinging on a gaussian potential barrier. Also multiple soliton states exist and there is a mutual interaction between the solitons if the phases of the different solitons are not equal. Single solitons and soliton trains have been recently observed experimentally with ^{7}Li atoms.

Quantum Rotation in Helium Clusters: a PIMC Study

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There have been many attempts to compute and predict the effective rotational constants of dopant molecules inside superfluid ^{4}He nanodroplets, which are experimentally observed to be smaller than the corresponding gas-phase values [1]. We have developed a method which allows us, in principle, to compute these effective rotational constants from first principles, and without restricting approximations. By treating the rotational degrees of freedom of dopants fully quantum mechanically, even very light rotors like HF can be studied.

The basis of our work is a Path-Integral Monte Carlo (PIMC) code by D. Ceperley *et al.* [2] that was modified in order to allow quantum rotation of dopants inside fairly large ($N \sim 300$) clusters of ^{4}He. The effective rotational constants of dopants are extracted from the imaginary-time correlation function of the molecular dipole, which is just the Laplace transform of the rotational spectrum.

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Intense pulsed helium droplet beams

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Pulsed (20-100 μs) nozzle beams have been used to generate helium droplets ($\langle \mathrm{N} \rangle = 10^4 - 10^5$). The dependence of the beam intensity and the mean droplet size on the source stagnation pressure and temperature are studied via mass spectroscopy and laser induced fluorescence of embedded phthalocyanine molecules. In comparison to a cw beam the pulsed source for the same pressure and temperature has a factor of 100 higher flux and the droplet sizes are an order of a magnitude larger.

Recent developments on superfluidity of Bose-Einstein condensates

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In this talk I will review the most recent experimental and theoretical results on the superfluid phenomena exhibited by inhomogeneous and finite-sized Bose-Einstein condensates. These include the propagation of Bogoliubov sound, quantized vortices, Josephson phenomena and, in general, the consequences of irrotationality.

Bubble formation and energy relaxation in electronically excited ^{3}He- and ^{4}He-clusters

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The energy relaxation and the transport features of ^{3}He- and ^{4}He-clusters are investigated using luminescence spectroscopy. He clusters do absorb light between 20 and 25 eV and are excited with monochromatic synchrotron radiation [1]. After excitation He clusters emit photons either in the visible and near infrared range (vis/ir) or in the vacuum ultraviolet spectral region. Because of the relatively high resolution dispersed luminescence spectroscopy in the vis/ir is well suited to investigate the population of the excited levels after electronic excitation of the clusters.

Depending on the cluster size and the isotopic constitution, either sharp, broadened or shifted emission bands of single electronically excited He molecules are observed [2]. Small and medium sized ^{3}He- and ^{4}He-clusters show a sharp line spectrum only. The sharp lines are un-shifted with respect to those of free He atoms and molecules and attributed to excited atoms and excimers which are desorbed within a short time interval after excitation.

Large ^{4}He-droplets also show shifted emission bands [3]. These bands are assigned to He molecules in bubbles because they resemble features which were observed in bulk liquid ^{4}He [4]. Bubbles, similar to those around electrons immersed in liquid He, are formed as a result of the repulsive Pauli force between the Rydberg electron of the electronically excited molecule and the closed-shell ground state He atoms. Larger ^{3}He droplets reveal features which are also assigned to electronically excited molecules enclosed in bubbles. However, these features are only broadened. In ^{3}He clusters bubbles are expected to be larger than in ^{4}He-clusters because the surface tension is lower. While bubbles appear only in very large ^{4}He droplets (10^7 atoms) they show up already in much smaller ^{3}He clusters (30000 atoms). This means that the dwell time of the bubbles is in ^{3}He- considerably longer than in ^{4}He-clusters. A lower average velocity is in line with diffusive motion due to the non-superfluid phase of ^{3}He clusters. One the other hand we assume that in ^{4}He droplets the molecules can move ballistically towards the surface. ¿From the intensity ratio of the luminescence of molecules which are either ejected or enclosed in bubbles, the cluster diameter and the radiative lifetime an average velocity of 7 m/s could be deduced.

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Alkali Clusters in Giant Spin States from Helium Nanodroplet Isolation

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Helium Nanodroplet isolation has been applied to form alkali clusters (Li_n, Na_n, K_n, Rb_n, Cs_n, $n \leq 20$). We use a beam of helium droplets (He_N, $N \approx 5000$) and consecutively pick-up several alkali atoms. Alkalis do not submerge in helium, in contrast to all other atoms and molecules probed so far. The atoms recombine on the surface of the droplet to form molecules and larger clusters. Like in the formation of alkali dimers and trimers on helium droplets [1], we find the larger clusters exclusively in high-spin states, i.e. the valence electrons are totally spin-polarized. In this way, non-metallic, van-der-Waals-like complexes of metal atoms are grown because of lacking strong bonds from pairing electrons. Species having paired spins efficiently desorb from the helium droplet because of their large recombination energy in comparison to the very weak binding to the surface of the droplets. We measured the abundance of cluster sizes by means of femtosecond multi-photon ionization and find sodium and potassium to readily form non-metallic clusters containing up to 20 atoms. In contrast, this process is suppressed for rubidium and cesium indicating a metal-insulator transition at very small cluster sizes (n=4, 6 for Rb, Cs, respectively). Apparently, for these alkalis, larger high-spin aggregates are not stable and depolarize upon cluster formation. Moreover, the measured mass distributions indicate that upon ionization by the femtosecond laser pulses, the high-spin systems collapse and metallic clusters are formed; the clusters fragment, heated by the large amount of binding energy released during this process.

Furthermore, laser-induced fluorescence is applied to study electronic properties of organic, semi-conducting nano-structures. Spectra of size-selected complexes of 3,4,9,10-perylene-tetracarboxylic dianhydride (PTCDA) are presented where the helium droplet environment enables us to separate charge transfer from molecular excitations. The results are discussed in comparison with spectra of PTCDA films and molecules probed in organic solvents.

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Structure and Rotation of Linear Molecules in Para-Hydrogen and Helium-4 Clusters

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We present a theoretical analysis of structure and spectroscopy of linear molecules in low temperature clusters of molecular hydrogen and helium-4 and mixed hydrogen/helium clusters. The ground state structures of para-H_2 and ^{4}He clusters around OCS are explored using diffusion Monte Carlo (DMC) simulations, and we obtain approximative rotational excitation energies with the clamped-coordinate quasi-adiabatic method (ccQA). In the case of $N = 1$, we compare to exact analytic results, and, in the case of $N > 1$, to exact stochastic results from the spectral evolution method (POITSE), which shows that the assumption underlying ccQA of a semi-rigid complex is valid for small clusters around OCS, $N \leq 5$.

As a microscopic approach complementary to stochastic methods, we employ correlated basis function (CBF) theory. We report about the formulation of the CBF theory for linear molecules in bulk quantum liquids, and results for the cases of small mass and large rotational constant (HCN) as well as large mass and small rotational constant (OCS). Furthermore, we outline the prospects for semi-analytic results from CBF apart from the calculation of effective rotational constants: using correlated basis wave functions in fixed node DMC and for the construction of projection operators in POITSE; the calculation of homogeneous line broadening (life-time); the calculation of microscopic currents patterns for the assessment of hydrodynamic models.

Structure and spectroscopy of OCS in Helium, Hydrogen and mixed Helium/Hydrogen clusters

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We present a comprehensive theoretical analysis of the spectroscopy of the OCS molecule in quantum clusters of helium and hydrogen, employing complementary finite-temperature path integral, and zero-temperature spectral evolution and diffusion Monte Carlo methodologies. Exact calculations of the rotational energy level constants of OCS in clusters of ^{4}He are shown to demonstrate a clear transition between a molecular complex and a quantum solvated molecule at a size smaller than one solvation shell [1]. The calculated rotational constants converge to the experimental value measured in large droplets, which was previously accounted for by the local two-fluid theory based on the path-integral Monte Carlo calculations for larger clusters [2]. A complete analysis of this size dependent behavior is given with the use of complementary rigid coupling approximations, and with the analysis of extent of adiabatic following of the helium solvent and of the onset of permutation exchanges among the helium atoms. The microscopic relation between direct energy level calculations and the local two-fluid theory for response of helium to molecular rotation is thereby clarified. Path integral calculations for OCS in hydrogen clusters show evidence of a low-temperature nanoscale superfluid state of molecular hydrogen that is characterized by a strong anisotropy and that can account for anomalous features in the infra-red spectra of OCS complexed by small numbers of hydrogen inside helium droplets [3]. The effect of the molecule-solvent interaction potential on the permutation exchange propensity of the solvating species is compared for the two species H_2 and He. The similarities and differences between the behavior of OCS in these two quantum clusters are further explored with calculations of the structural and spectral properties of OCS embedded in mixed complexes of helium and hydrogen.

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Helium atom "transmission" through a slab of liquid ^{4}He

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We have measured the "transmission" of helium atoms through a thin slab of liquid ^{4}He. What is actually happening is that helium atoms quantum condense on the surface of the liquid and create rotons, one atom creating one roton. The rotons propagate through the liquid and quantum evaporate atoms from the far surface of the liquid, again one roton creates one atom. If the surfaces of the liquid are curved then there is strong refraction and some rotons can hit the side of the glass cylinder which is holding the liquid helium. If this happens the rotons are lost and do not cause evaporation.

The atom beam is created by a heat pulse in a thin metal film covered in layer of liquid helium and the atoms are detected by a superconducting Zn bolometer. The thin slab of liquid helium is contained in the parallel pores of a glass plate; the pores are 50 microns diameter and the plate 200 microns thick. The curvature of the surfaces are controlled by the chemical potential of the helium. The temperature is less than 70mK so the atom and roton mean free paths are long. We have measured the atom transmission as a function of the curvature of the liquid's surface and compared it to modelled results. The transmission, when the surfaces are flat, is compared to the microscopic theory of Campbell, Krotscheck and Saarela [1].

[1] C.E. Campbell, E. Krotscheck and M. Saarela, J. Low Temp. Physics **113**, 519 (1998).

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