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Hydrogen atoms stored near absolute zero neither form molecules nor condense into a liquid. Instead, they are part of the intriguing world of quantum gases

Atomic hydrogen: the quantum gas

JOOK WALRAVEN

IT WAS a complete surprise when Isaac Silvera and his colleagues at the University of Amsterdam observed the existence of a remarkably stable gas of hydrogen atoms in 1979. They found that a gas of the so-called "atom of atoms" could be confined for several minutes in a cryogenic test cell close to absolute zero, provided that the walls were coated with a film of liquid helium. Until then, if researchers wanted to study monoatomic hydrogen for any length of time they had to use a discharge source to replace those hydrogen atoms that recombined to form molecular hydrogen. But most important, Silvera's experiments marked the first experimental investigation of a dilute quantum gas.

At the time the gas phase of hydrogen was regarded as an interesting anomaly, unique from many points of view, but not representative of a general class of experimentally accessible systems. However, over the last decade this perspective has changed entirely. Today, thanks to the development of optical cooling methods, all sorts of atoms can be stored as gases at very low temperatures. One technique is the "magnetic bottle", in which a configuration of magnetic fields traps a cloud of magnetic atoms in an effective potential well, without the need for any surfaces to confine the gas. The trapping forces in these bottles are small, meaning that the trapped atoms are ultracold – typically below 1 mK.

Ultracold gases that have been trapped in magnetic bottles include wildly different systems, such as the heavy alkalis rubidium (Rb) and caesium (Cs), and the metastable triplet helium (He^*). Given the range of atoms that can be obtained in gaseous form at very low temperatures, two interesting questions need to be asked. First, what turns an ultracold gas into a quantum gas? And second, what is the position of hydrogen among the ultracold gases?

Charles Hecht of the University of Chicago first conjectured the idea of a gaseous phase of hydrogen atoms in 1959, but it wasn't until 1973 that Richard Etters at Colorado State University gave the first convincing prediction of such a gas. Since then the properties of the atomic hydrogen gas have been studied extensively. Many valuable theoretical insights into the unique nature and stability of hydrogen – and other ultracold gases as well – have come from Yuri Kagan's and Georgi Shlyapnikov's groups at the Kurchatov Institute in Moscow, and from Boudewijn Verhaar and colleagues at Eindhoven University of Technology.

Quantum gases, even before they were detected, were also routinely used in university courses to teach many-body behaviour in low-temperature physics and to introduce phenomena such as superfluidity and superconductivity. For example, when Landau and Lifshitz considered "imaginary" quantum gases they wrote: "The problem of the thermodynamic properties of an almost ideal, highly degenerate gas has no direct physical significance, since gases which actually exist in nature condense at temperatures near absolute zero." In other words, if gases could be cooled to the low temperatures needed to observe quantum behaviour, they would not remain gaseous. Indeed, the stable low-temperature phase of hydrogen is no exception – it forms the well known molecular quantum solid H_2 (p43).

However, if ultracold gases are metastable – that is unable to reach an equilibrium condensed state – they exhibit the properties of a quantum gas. This metastability is analagous to the way an undercooled rain droplet can, for some time, find it hard to freeze. A gas of hydrogen atoms represents the mildest form of metastability, as it is the least likely to condense. The system will only condense when three atoms collide simultaneously, an extremely rare process under certain conditions. The most extreme form of metastability in ultracold atoms is found in matter-wave resonators – cavities for quantum mechanical matter waves (similar to optical resonators) – in which the strict absence of any collision is of paramont importance.

To distinguish between the various types of ultracold gases there are three indicators that show when a gas is behaving as a quantum gas. These are the zero-point motion of atoms, non-classical scattering effects when two atoms collide, and quantum degeneracy.

The energy balance near absolute zero

When physicists want to compare various substances, the proven method is corresponding states analysis. This approach, which can be traced back to the work of Johannes Diderik van der Waals at the start of this century, is based on the observation that the interaction potential between two atoms always has a similar shape – approximated, for example, by the Lennard-Jones potential. For classical substances, corresponding states analysis provides a unifying description for all substances in terms of a single set of reduced thermodynamic variables. It means that the thermodynamical phase in one substance at a given temperature and pressure has a corresponding phase with similar properties in another substance at a different temperature and pressure.

This approach was extended to quantum mechanical systems by Jan de Boer at the University of Amsterdam after the Second World War. De Boer introduced a quantum parameter, η, to give a measure of the importance of zero-point motion – the motion a substance has at absolute zero (figure 1): $\eta = \hbar^2/mr_o^2\varepsilon$, where $\hbar$ is Planck's constant divided by 2π, r_o is the hard core radius of the interaction potential between two atoms of mass m, and ε is the potential well depth.

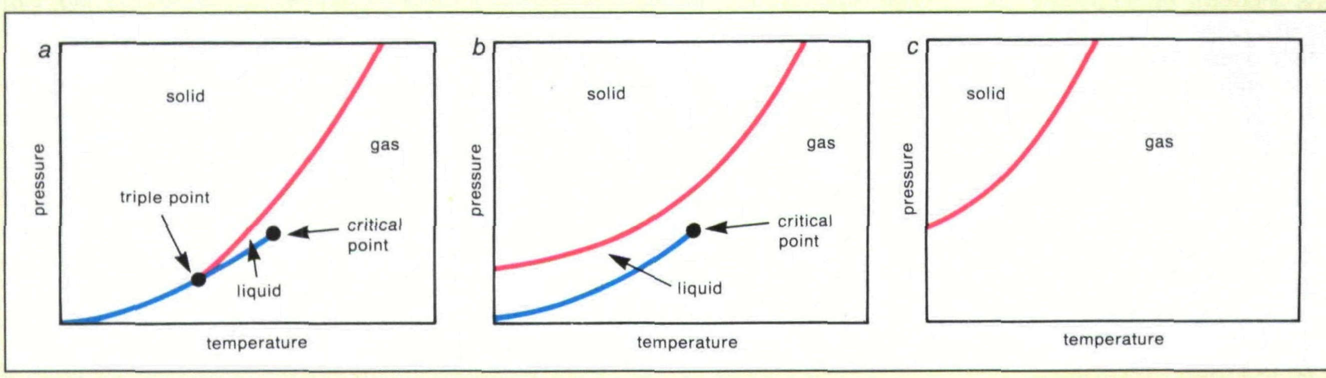
The corresponding states analysis at, or near, absolute zero is simple, because the potential and kinetic energies of only the ground state need to be considered. At this temperature the interatomic potential, which is attractive (except at very short interatomic distances), tends to encourage atoms to bind together. However, the confinement of atoms by neighbouring atoms, which gives rise to zero-point motion, encourages repulsion.

The simple qualitative rule for quantum gas behaviour is that the repulsion between two atoms should outweigh the attraction. This means that the kinetic energy in a quantum gas should be larger than the potential energy, thereby making the ground-state energy, E_o, positive. The cohesive forces are then too weak to bind the atoms into a solid or a liquid. In other words the quantum gas lacks a many-body bound state.

In 1975 Lewis Nosanow of the US National Science Foundation and Michael Miller of the University of Massachusetts at Amherst used corresponding states to provide a quantitative guide to predict which systems become quantum gases. They calculated that if $\eta>0.46$ for bosons (particles with integer spin) and $\eta>0.29$ for fermions (particles with half integer spin) then the substance is a true quantum gas. The table below shows that hydrogen is the only substance whose atoms satisfy this criterion. All other systems tend to form solids or, in the case of ^3He and ^4He, quantum liquids at sufficiently low temperatures (figure 1). As will be shown later, this feature makes the metastability of hydrogen atoms different from that of any other ultracold gas.

Quantum parameters for various substances

Substance	H	D	^{3}He	^{4}He	H_2	Li	Na	Cs
η	0.55	0.275	0.24	0.18	0.076	0.0015	5×10^{-4}	3×10^{-5}



1 Typical phase diagrams of matter at increasing degrees of quantum behaviour, defined by the values of the quantum parameter η. (*a*) At small η the system behaves classically: it is solid at absolute zero, but can form a liquid or gas at higher temperatures, and has both a triple point and a critical point. (*b*) At intermediate η, where the system shows signs of quantum behaviour, it has a liquid "ground state" at absolute zero, can also form a solid or a gas, but has no triple point. (*c*) At large η the system has a gaseous ground state, but no triple point and no critical point. Because the highly quantum system shown in (*c*) is metastable, even the solid pocket at high pressure is unlikely ever to be observed.

Collide and scatter

Large atoms, such as rubidium and caesium, can also be quantum gases, even though they fail to satisfy the corresponding states criterion mentioned above. To understand why, one has to consider the quantum mechanical effects that take place when pairs of atoms collide.

The extent to which an atom in an ultracold gas is localized in space is given by the thermal de Broglie wavelength, $\Lambda = (2\pi\hbar^2/mkT)^{1/2}$, where k is Boltzmann's constant and T is the temperature. If Λ is less than the range of interatomic interaction, R_o, the atoms behave as classical, point-like particles, but if Λ is much greater than R_o, the interaction between two atoms will be affected by quantum mechanical interference.

For a light atom such as hydrogen, which has a small R_o, this condition for quantum behaviour is met at a temperature as high as 1 K. However, for caesium, which has a very large R_o, quantum behaviour only shows up at much lower temperatures – typically below 30 μK. Therefore the de Broglie wavelength, rather than the temperature, defines the regime of quantum behaviour in ultracold gases.

There are also big differences between the way bosons and fermions behave. Two atoms separated by a distance r have a centrifugal energy $E_r = l(l+1)h^2/mr^2$, where l is the quantum number of the angular momentum of relative motion. If the two atoms are to scatter – and thus exhibit quantum behaviour – they have to approach to within R_o. When $l=0$ the atoms can do this, but when $l>0$ there is a centrifugal energy barrier that prevents the atoms from approaching one another (figure 2).

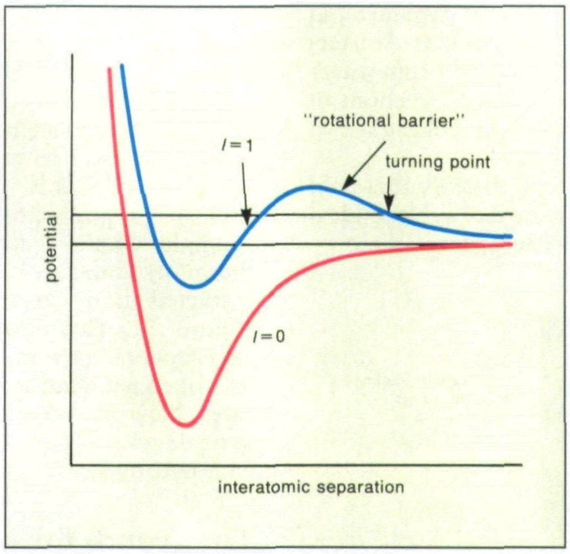
Fermions, which have an odd l, cannot penetrate the centrifugal barrier, so they hardly scatter at all. However, bosons, which have an even l, can approach each other and scatter. Almost all boson scattering therefore takes place in collisions where $l=0$, known as *s*-wave collisions.

In a dilute gas, where atoms are generally far apart, atoms only interact via simple binary collisions. For a gas of ultracold bosons, these collisions are isotropic, and the amplitude (or "length") of the *s*-wave scattering depends on the shape of the potential energy curve that governs the interatomic interaction. If the scattering length is positive, the overall effect when two atoms are involved in a binary collision is that they repel one another; but if the length is negative, the overall effect is an attraction. The scattering cross-section is, however, always positive because it depends on the square of the amplitude.

In a boson gas of this type the ground-state energy per atom, E_o, is linearly dependent on both the density of atoms and their scattering length. Thus, in contrast to the corresponding states prediction for quantum gas behaviour – which said only light atoms such as hydrogen could satisfy the $E_o>0$ condition – heavy atoms can also have $E_o>0$, provided their scattering length is positive. Therefore any substance with positive scattering length at sufficiently low densities can form a quantum gas. This assumes, of course, that the density fluctuations in heavy-atom gases that give rise to triple, quadruple or higher order collisions – and which would eventually lead to the formation of condensed matter – can be neglected. This assumption is certainly true on short timescales.

The case of a negative scattering length is special. This system wants to lower its energy by contracting, but cannot do so if the energy liberated cannot be carried away – by escaping atoms, for instance.

A stream of papers has started to appear in the scientific literature, the authors of which either calculate the sign of the scattering length of various substances or discuss how the sign is affected by the presence of externally applied fields. Hydrogen is the only system for which the scattering length can be calculated from first principles. (All other systems depend on input from experiments.) Hydrogen atoms have an anomalously small scattering length – 0.07 nm – only slightly larger than the Bohr radius. This makes hydrogen atoms the most weakly interacting boson gas to have been investigated experimentally.



2 The presence of a "rotational barrier" between two atoms with a relative rotational quantum number $l > 0$ means that the atoms cannot approach each other at low temperatures because there is not the energy available to cross the barrier. The atoms can, however, approach each other if $l = 0$.

Flipping spin

Heavy atoms, such as caesium, can only exist as quantum gases under metastable conditions at low densities ($< 10^{15}$ cm^{-3}), because if three or more caesium atoms collide to form dimers – through processes such as Cs + Cs + Cs → Cs_2 + Cs – a true equilibrium many-body bound state can eventually be reached. The equilibrium state will either be a liquid or a solid, depending on the value of the quantum parameter, η.

Ultracold hydrogen is very different. Its metastability depends on the spin state of the hydrogen electron, which can be either parallel to the applied field – spin up (H↑) – or antiparallel – spin down (H↓). This spin polarization is achieved experimentally by applying a magnetic field gradient across an unpolarized gas. H↑ collects at the low-field end.

For spin-polarized hydrogen, in which the spins of the electrons in different atoms are all parallel, the interaction between two hydrogen atoms is very weak, and a quantum parameter of η = 0.55 reflects this. For antiparallel spins the interaction between two hydrogen atoms is strong, leading to the formation of an H_2 molecule.

The spin of hydrogen electrons can, however, be "flipped" from H↓ to H↑ by interatomic forces. The cohesive van der Waals force between two hydrogen atoms does not have the proper symmetry to induce a spin flip, but the magnetic interaction between the spins – known as the spin dipole interaction – does, even though it is a thousand times weaker than the van der Waals force. Spin-polarized hydrogen atoms are therefore stable under the influence of van der Waals forces, but the spin dipole interaction can lead to the formation of the H_2 molecule.

For spin-down polarized hydrogen, studied in a sample cell covered with liquid helium, spin relaxation is practically impossible in the binary collision H↓ + H↓ → H↑ + H↑. This is because the activation energy needed to turn the spin against the magnetic field cannot be supplied by the thermal motion of the ultracold atoms. Spin flips are only possible in a dipole induced direct three-body recombination process, H↓ + H↓ + H↓ → H_2 + H↑ (or H↓). Here the energy liberated in the formation of the molecular bound state enables the reaction to take place.

However, because the spin-dipole interaction is so much weaker than the van der Waals interaction, the spin dipole-induced recombination rate in H↓ turns out to be very slow. Indeed, an H↓ atom can survive 10^{10} times as long as a caesium atom. Also, because H↓ recombination is a three-body collision process, the rate is proportional to the square of the density. This means that an H↓ gas can be 10^5 times as dense as a caesium gas, yet survive for the same length of time. Ultracold H↓ exemplifies the mildest form of metastability, allowing densities comparable with the earth's atmosphere (~2×10^{19} cm^{-3}) to be investigated.

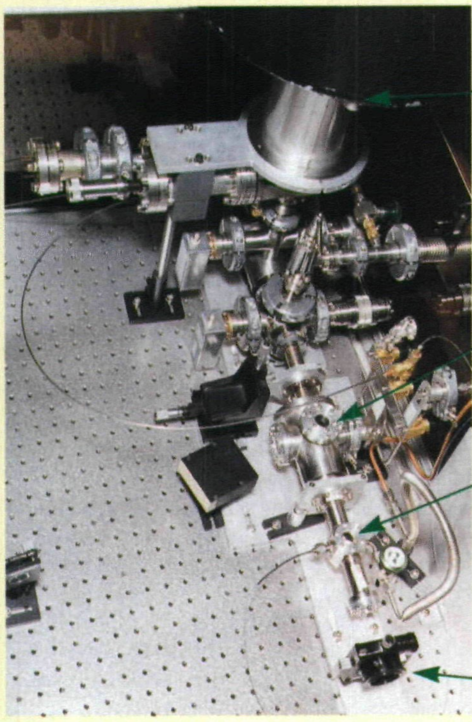
For spin-up polarized hydrogen, H↑, obtained by confinement in a magnetic bottle, the situation is totally different. H↑ is much less stable than H↓, because spin-dipole relaxation can indeed occur in a binary collision, H↑ + H↑ → H↓ + H↓. The process is exothermic and proceeds without thermal activation. The spin-flipped atoms are then ejected from the trap.

It is interesting to note that spin-dipole relaxation does not impose a fundamental limit on the densities that can be achieved with H↑. Although the H↑ gas may survive longer at lower densities, it also takes longer to establish thermal equilibrium. Therefore the highest density that can be studied is limited by the minimum time required to make a meaningful measurement.

Evaporative cooling

Hydrogen is the model ultracold quantum gas because it can be used to compare experimental information with accurate theoretical predictions. The bosonic nature of hydrogen atoms, as identified from the effects of binary collisions, has been observed in nuclear spin dynamics, in nuclear magnetic relaxation by Ad Lagendijk at Amsterdam in 1984 and, more dramatically, in nuclear spin waves – collective excitations of the spin degrees of freedom of the gas – by Jack Freed, David Lee and colleagues at Cornell University in 1984.

In Amsterdam, Jaap Berkhout showed in 1989 that cold hydrogen atoms near a liquid helium surface are the ideal system for investigating the ultra-low energy limit of atom–surface scattering. This could also be used for testing the quantum theory of physisorption, and for observing quantum reflection at vanishing energies. These experiments were extended to sub-milliKelvin temperatures by Thomas Greytak and Daniel Kleppner at MIT in 1993. The highest atomic hydrogen density – close to the density of the Earth's atmosphere – was observed by Simo Jaakkola at the University of Turku, Finland, in 1993.



3 Advanced optics and advanced cryogenics come together in this apparatus used by Amsterdam University to study a gas of hydrogen atoms in a magnetic trap. The tripling cell lowers the wavelength of the 365 nm incoming pump beam by a third to 121 nm. This radiation then probes the atomic hydrogen in the trap at a temperature close to absolute zero.

The hydrogen atom was also the third neutral particle to be confined in a neutral particle trap, after the neutron in 1978 and sodium in 1985. Harold Hess at MIT proposed a cryogenic filling method in 1986 that could load H↑ in a magnetic trap, exploiting the advantage that atomic hydrogen, unlike other ultracold gases, can be handled with cryogenic methods. High densities were reached in these traps, first at MIT in 1987 and then at Amsterdam in 1988.

This work developed the concept of evaporative cooling, an extremely efficient cooling mechanism that had been proposed by Hess in 1986. In evaporative cooling, the most energetic atoms resulting from interatomic collisions escape from the trap and cool the gas – just as escaping hot vapour atoms cool a cup of tea. The temperature of the gas is then established by measuring the energy distribution of the atoms after they escape. This approach allowed John Doyle and colleagues to reduce the hydrogen gas temperature by three orders of magnitude to a record value of 100 μK at MIT in 1991.

In Amsterdam, optical cooling and thermometry methods were developed for trapped hydrogen atoms by Jom Luiten and Irwan Setija in 1993 (see "Laser cooling the atom of atoms" by Dieter Meschede *Physics World* June 1993 pp25–26). These experiments are the only example where optical cooling can be regarded as a thermodynamic cooling mechanism by which heat is extracted more slowly than the interatomic collision rate (figure 3). Remarkably, it is more difficult to find experimental techniques to observe an ultracold atomic hydrogen gas than it is to reach sub-milliKelvin temperatures. New and more sensitive detection schemes will have to be developed before lower temperatures can be achieved with hydrogen.

The search for quantum degeneracy

The quantum mechanical degeneracy effects mentioned in the quote from Landau and Lifshitz are among the most intriguing physical phenomena of ultracold gases that have yet to be observed. When a pair of ultracold atoms are closer than their de Broglie wavelength, quantum mechanical interference effects influence the detailed behaviour of these atoms. These quantum correlations are significantly different for bosons and fermions, and they manifest themselves whether or not there is any interatomic interaction.

Most importantly, quantum correlations affect the statistical distribution over the available free particle states. There is no restriction on how many bosons can occupy a given state, but no more than one fermion can be in the same state. At low gas densities these restrictions are of little consequence and the deviations from classical (Boltzmann) statistics remain small. However, as soon as the mean separation between atoms of density n becomes comparable to, or smaller than, the thermal de Broglie wavelength, $n\Lambda^3>1$, the difference between boson and fermion statistics becomes dramatic.

The fermions become entangled in complicated correlated motions. As the density increases, all thermally accessible states gradually become occupied, leaving no phase space for scattering. Ultimately, when the interatomic interaction is important, Cooper pairing and superfluidity can occur. In view of the present knowledge of the metastability of ultracold gases, it seems inconceivable that the densities required to observe superfluidity in ultracold fermion gases will ever be reached.

Bosons, however, are very different. Dilute boson gases behave more or less classically until the degeneracy parameter reaches a value $n\Lambda^3=2.612$. At this point the statistics favour the massive occupation of the ground state in a phase transition known as Bose-Einstein condensation (BEC). In an interacting boson gas, BEC would be accompanied by a superfluid transition similar to that observed in liquid ^{4}He. Remarkably, the interactions in the Bose condensed state can play a very prominent role, despite the very dilute condition of the parent gas, and even at temperatures much higher than those associated with the effects of the interatomic interactions. This is particularly true for inhomogeneous systems, such as the magnetically trapped gases, in which the Bose condensate is expected to be highly compressed (in contrast with the condensate in liquid ^{4}He).

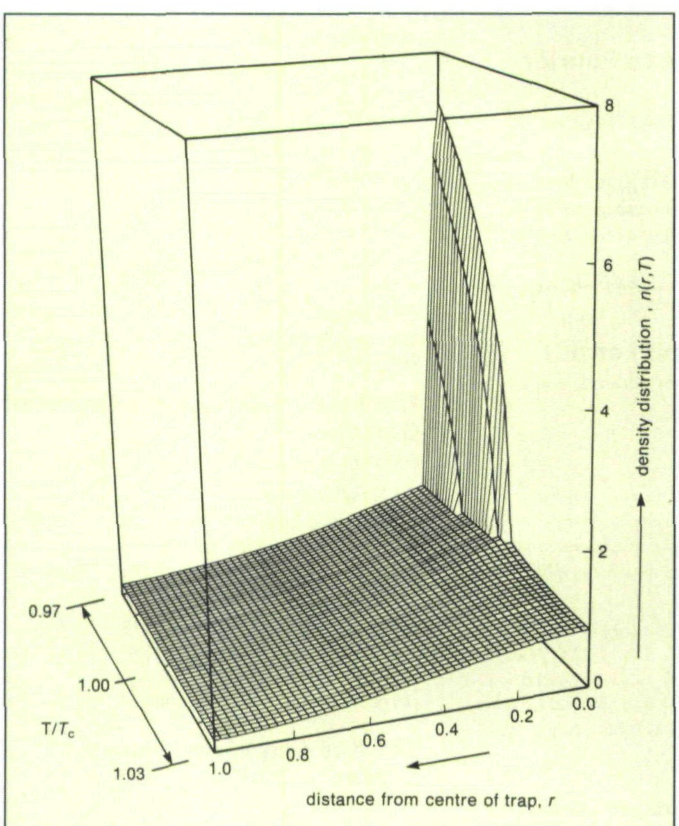
This compressed Bose condensate can be identified by a peak in the density distribution at a certain position in the trap holding the condensate (figure 4). The search for and characterization of this peak – both in real and momentum space – is one of the major challenges of BEC experiments on ultracold gases. So far, degeneracy parameters within a factor of 10 of the BEC limit have been studied. This gives us confidence that, if a little extra effort is put in, BEC can be observed in these dilute gases.

Hydrogen is a prominent candidate for observing BEC, and improved detection methods are currently under development. However, very rapid progress has been made this year with the ultracold alkali gases, notably by Randall Hulet at Rice University working on lithium, Wolfgang Ketterle at MIT on sodium, and Eric Cornell and Carl Weiman at the Joint Institute for Laboratory Astrophysics in Boulder, Colorado, on rubidium.

The atom laser – a metastable future?

Finally, I should make some remarks on the idea of the "boser" – an "atom laser" that produces a coherent beam of degenerate bosons. The boser is based, in close analogy to the optical laser, on the multiple occupation of selected high-lying modes in a matter wave resonator. However, many interesting problems have yet to be solved before a boser can be built. These include the population of the mode, the build-up of coherence and how to create the output – concepts that push the relevance of metastability to its extreme.

Also, while Bose-Einstein condensation is concerned with the occupation of the ground state under the influence of Bose correlations and collisions in a thermal system, the Bose correlations in an atom laser should do their work in the absence of collisions and with a non-thermal bath – for example by a cleverly chosen electromagnetic field. Certainly, very interesting times lie ahead as we wait to see how these fragile ideas develop in the future.



4 Density distribution of a gas of hydrogen atoms, calculated for a parabolic trapping potential well, where r is the distance from the centre of the trap. At temperatures above the onset temperature for Bose-Einstein condensation (T_c), where the system behaves classically, the density distribution is smooth. However, as soon as the temperature drops to even a few per cent below T_c, a pronounced peak in the density distribution develops. This indicates that Bose-Einstein condensation, identified by a massive occupation of the ground state, has taken place.

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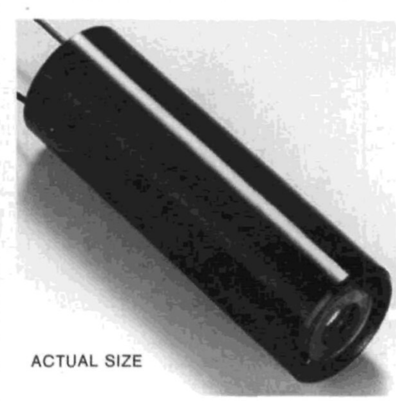

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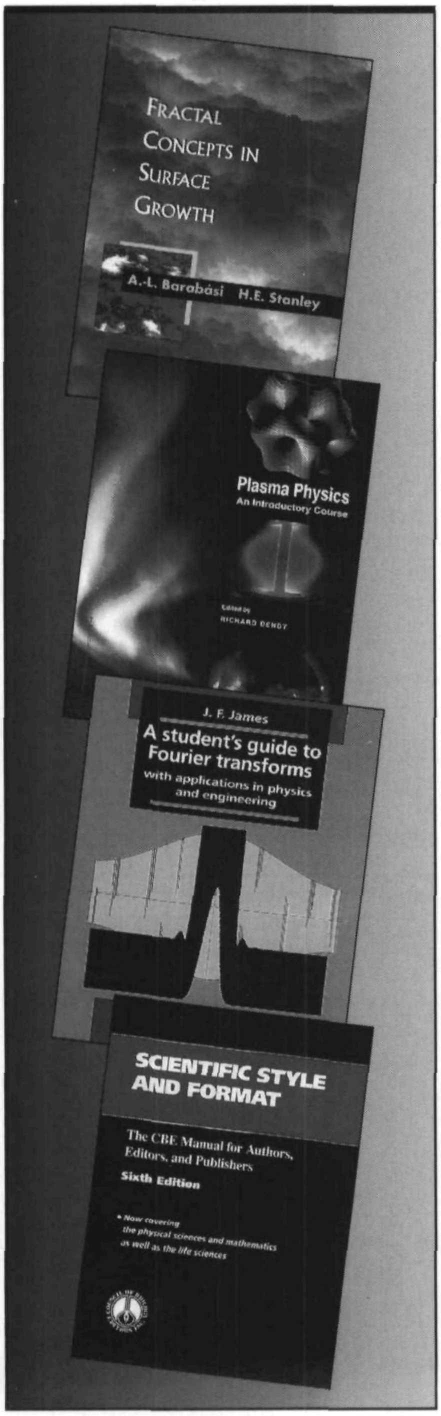

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