Towards the elusive Efimov state of the $^4$He$_3$ molecule through a new atom-optical state-selection technique

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Excited states and excitation energies of weakly bound systems, e. g. atomic few-body systems and clusters, are difficult to study experimentally. For this purpose we propose a new and very general atom-optical method which is based on inelastic diffraction from transmission gratings. The technique is applicable to the recently found helium trimer molecule $^4$He$_3$, allowing for the first time an investigation of the possible existence of an excited trimer state and determination of its excitation energy. This would be of fundamental importance for the famous Efimov effect.

Already in 1935 Thomas \cite{1} discovered a surprising property of three-body systems. He considered short-range two-particle potentials which supported just one single bound state with an arbitrarily weak binding energy. He then found that for the three-body system there could exist a much more tightly bound ground state and that the binding energy could approach $-\infty$ when the range of the two-particle potential approached zero. Thirty-five years later Efimov \cite{2} obtained a striking generalization of this result. He predicted that when one weakened the two-body interaction the number of three-body excited states could increase to infinity. An excited state which appears under weakening of the two-particle potential is called an Efimov state. Conversely, under strengthening of the two-particle potential an Efimov state disappears into the continuum. Intuitively one can understand the Efimov effect in a three-body system by imagining a weakly bound, and therefore spatially very extended, two-particle subsystem. This subsystem can then act on the third particle with a force whose range is given by the spatial extent of the subsystem \cite{3}. This range therefore increases more and more when the two-particle potential is decreased. As opposed to a short-range potential \cite{4}, however, a long-range potential can have infinitely many excited states.

Whether the Efimov effect does occur in nature is still an open question. In nuclear physics no generally accepted examples of Efimov states have been found \cite{5}. For systems of neutral atoms, however, they might exist, and an excellent candidate is the helium trimer, $^4$He$_3$, since the dimer, $^4$He$_2$, is believed to have an extremely weak binding energy ($\approx -1.3$ mK) and no excited states. A few years ago $^4$He$_3$ has been observed by Luo et al. \cite{6} and independently by Schöllkopf and Toennies \cite{7}, who also observed $^4$He$_3$. There has been a lot of theoretical work on the existence of an Efimov state in the helium trimer, with sometimes conflicting results \cite{8,9,5,10,11,12}. The overall theoretical picture presently indicates the existence of a ground state and a single excited state, denoted here by $^4$He$_3$ and $^4$He$_3^*$, respectively. The latter is believed to be an Efimov state. Both are $s$ states with respective energy around $E_0 = -0.1$ K and $E_1 = -2$ mK \cite{13}. Experimentally, $^4$He$_3^*$ has not yet been seen \cite{14}.

The Efimov property as well as the very existence of the excited state $^4$He$_3^*$ depend sensitively on the detailed form of the two-atom interaction, and even small retardation corrections to the potential can significantly affect the $^4$He$_2$ binding energy and bond length \cite{13}. A precise knowledge of this potential is also necessary for understanding liquid helium droplets \cite{14} and superfluidity. Therefore conclusive experimental evidence of an excited state $^4$He$_3^*$ and determination of its binding energy would not only be a crucial step towards establishing the existence of an Efimov state but would also give important information on the He-He potential.

Here we propose a new and very general method to both detect and select an excited state of $^4$He$_3$, or of other systems, as well as to determine its excitation energy. Excitation can be achieved by scattering a beam from a solid surface \cite{15} or, in our case, from many small surfaces. Taking for the latter the bars of a microfabricated transmission grating \cite{16} one can achieve excitation and separation at the same time, as will now be shown.

The state-selective property stems from two conservation laws. If the incident molecular center of mass momentum is denoted by $P'$ and the final one by $P$, energy conservation implies \cite{19}

$$P'^2/2M = P^2/2M + \Delta E_{\text{int}}$$

(1)

where $M$ is the molecular mass and $\Delta E_{\text{int}}$ accounts for a possible change of the internal molecular state. The other conservation law comes from the discrete translational invariance of the grating in the 2 direction (period $d$, cf. Fig. \cite{16}). This implies the conservation of the initial momentum component $P'_2$ up to a reciprocal lattice vector \cite{20}, i. e.

$$P_2 = P'_2 + n2\pi\hbar/d$$

(2)
where \( n = 0, \pm 1, \pm 2, \ldots \). With \( P_2 = P \sin \varphi \) (see Fig. 1) this yields a relation between the angle of incidence \( \varphi' \) and the allowed angle, \( \varphi_n \), of the \( n \)-th order diffraction peak. With \( \lambda' = 2\pi h/P' \) and \( \lambda = 2\pi h/P \) the initial and final de Broglie wave length, the relation can be written, after a short calculation, as

\[
\sin \varphi_n = \frac{\lambda}{\lambda'} \sin \varphi' + n\frac{\lambda}{d}.
\]

(3)

Eq. (3) holds for any molecule-grating interaction potential. A wave theoretical interpretation of Eq. (3) can be given as follows (see Fig. 1). First, a wave with angle of incidence \( \varphi' \) is refracted in the plane \( A \), with a change of wave length from \( \lambda' \) to \( \lambda \). The refraction angle \( \varphi_0 \) and the incidence angle \( \varphi' \) are related as in Snell’s law (21) through \( \sin \varphi_0/\sin \varphi' = \lambda/\lambda' \). Secondly, in the plane \( B \), the new wave of wave length \( \lambda \) is diffracted by the slits as in classical optics (22). Combining this with Snell’s law gives Eq. (3) (23).

For an elastic process \( \lambda \) coincides with the initial \( \lambda' \) and then Eq. (3) reduces to the condition for grating diffraction of a de Broglie wave as obtained from classical optics (22). But if the molecule is excited by the interaction with the grating, \( \lambda \) differs from \( \lambda' \) by a factor of \( 1/\sqrt{1 - (E_1 - E_0)/E_{\text{kin}}' \approx 1 + (E_1 - E_0)/2E_{\text{kin}}'} \) where \( E_{\text{kin}}' \equiv P'^2/2M \) is the initial kinetic energy. For \( ^4\text{He}_3 \) three different kinds of processes can occur, namely elastic scattering (\( ^4\text{He}_3 \rightarrow ^4\text{He}_3 \)), excitation (\( ^4\text{He}_3 \rightarrow ^4\text{He}_3^* \)) and breakups (\( ^4\text{He}_3 \rightarrow ^4\text{He}_2 + ^4\text{He} \) or \( ^4\text{He}_3 \rightarrow ^4\text{He} + ^4\text{He} \)) where the latter have diffuse scattering angles. For the elastic case (\( \lambda = \lambda' \)) the diffraction term \( n\lambda/d \) in Eq. (3) has been used previously to separate molecules of different mass (22).

In order to separate the equal mass particles \( ^4\text{He}_3 \) and \( ^4\text{He}_3^* \) we propose here to use the first term in Eq. (3) (Snell’s law) and its dependence on the incidence angle as follows. For normal incidence of \( ^4\text{He}_3 \) the low order diffraction-peak angles of \( ^4\text{He}_3 \) and \( ^4\text{He}_3^* \) differ by micro radians only and are practically indistinguishable (\( \varphi' = 0 \) in Eq. (3)), but by rotating the grating (\( \varphi' \neq 0 \)) the peaks will separate. This allows the identification of \( ^4\text{He}_3^* \) and determination of \( E_1 - E_0 \). For example, the zeroth order \( ^4\text{He}_3^* \) diffraction angle gives

\[
E_1 - E_0 = \frac{P'^2}{2M} \left( 1 - \frac{\sin^2 \varphi'}{\sin^2 \varphi_0} \right).
\]

(4)

To quantitatively check the feasibility of our proposal we have calculated diffraction patterns of a \( ^4\text{He}_3 \) beam (\( ^4\text{He}_3 \rightarrow ^4\text{He}_3^* \), \( ^4\text{He}_3 \rightarrow ^4\text{He}_3 \)) incident under various angles on a 100 nm period silicon nitride (SiNx) transmission grating and for a typical nozzle temperature of 6 K (24,26,27). For this we have applied the approach of Refs. (24) to an incident three-body system with the trimer wave functions of Fig. 3. The wave functions have been obtained by means of the momentum space Faddeev approach and the unitary pole approximation (see e. g. Ref. [29]) using the Tang, Toennies, Yin (TTY) potential (29) and they are sufficiently accurate to yield \( E_0 = -0.1 \) K and \( E_1 = -2.1 \) mK, comparable to the results of the adiabatic hyperspherical approach in Ref. (10). In the calculation of the diffraction pattern in the Fraunhofer regime the trapezoidal cross section of the grating bars with a wedge angle of \( \beta = 9^\circ \) (see Fig. 4) and the helium-silicon nitride van der Waals interaction potential of Ref. (30) have been included. As can be seen in Fig. 3 a \( ^4\text{He}_3^* \) signal appears in the form of side peaks on the elastic diffraction peaks for angles of incidence \( \varphi' > 10^\circ \). The energy difference \( E_1 - E_0 \) is very small compared to the initial kinetic energy and results in small angle differences between \( ^4\text{He}_3 \) and \( ^4\text{He}_3^* \) diffraction peaks. But such small angle differences are easily resolvable in present-day experiments.

Fig. 3 reveals the role played by the attractive van der Waals interaction between the molecules and the grating material. While the elastic processes qualitatively follow the predictions of classical optics, the total transmitted \( ^4\text{He}_3^* \) intensity (\( ^4\text{He}_3 \rightarrow ^4\text{He}_3^* \)) is only slowly varying over a wide range of incidence angles \( \varphi' \). Therefore, the excitation of the molecules depends only weakly on the slit projection orthogonal to the incidence direction and will take place mainly at the slit boundaries. If the angle of incidence approaches the wedge angle of the grating bars (see Fig. 4) the \( ^4\text{He}_3^* \) transmission probability reaches its maximum value. In this case of grazing incidence on the surface of one side of the bars the van der Waals interaction strongly affects the excitation process and leads to a gain in the total transmitted \( ^4\text{He}_3^* \) intensity. Theory shows the excitation process to be the more efficient the larger the overlap region of the two wave functions.

Fig. 3 indicates that a realization of the proposed experiment requires the measurement of intensities over five orders of magnitude. With conventional electron impact ionization mass spectrometer detectors this accuracy has been achieved so far solely for helium-atom beams (34) because the estimated detection efficiency is only \( 10^{-8} \). A helium-atom detector with an efficiency of about \( 10^{-8} \) – \( 10^{-9} \) might be a useful spectroscopic tool for the investigation of internal atomic and molecular processes in these excited states.
molecular transitions which are not easily accessible to
laser light.

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FIG. 1. Wave theoretical interpretation for the selection of excited molecules.

FIG. 2. (a) Hypperradial probability density of the $^4$He$_3$ ground (solid line) and excited state (dashed line), and (b) radial probability density of the $^4$He$_2$ bound state (solid line) and TTY potential (dotted line). These states show the peculiar property that a substantial fraction of the probability densities is located far outside the potential well.
FIG. 3. Diffraction intensities for elastic ($^4\text{He}_3 \rightarrow ^4\text{He}_3$) and excitation ($^4\text{He}_3 \rightarrow ^4\text{He}_3^*$) processes of a ground state helium trimer beam at a nozzle temperature of 6 K, incident on a 100 nm period silicon nitride transmission grating under different angles: (a) $\phi' = 0^\circ$, (b) $\phi' = 10^\circ$, (c) $\phi' = 20^\circ$ and (d) $\phi' = 30^\circ$.

FIG. 4. Total transmission probabilities of elastic and excitation processes for a ground state helium trimer beam diffracted from a 100 nm period silicon nitride transmission grating at nozzle temperatures of 6 K and 30 K as functions of the angle of incidence. The dashed line is the transmission curve obtained from geometrical optics.
