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**Editorial for whichever issue of Molecular Physics
includes TMPH 219660, the paper by Professor Leif
Holmlid.**

Tim Softley

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**Editorial for whichever issue of Molecular Physics includes
TMPH 219660, the paper by Professor Leif Holmlid.**

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Editorial: Rydberg Matter

In this issue we publish a paper by L. Holmlid which reports the observation of radiofrequency emission spectra that are attributed to rotational transitions of planar Rydberg matter clusters K^*_{19} . This work extends the wide-ranging observations of such clusters made by Holmlid and his coworkers published in over 100 papers in the last 10 years. The clusters are reported to be formed via thermal desorption of electronically excited atoms from a K-promoted iron oxide catalyst material at temperatures in the range 300 - 900K. As in previous work [2,3], it is believed that these electronically excited atoms condense into the electronically excited clusters, exchanging some excess energy with the solid at the surface boundary layer. The structure of the clusters K^*_N , where N is one of the magic numbers 7, 19, 37, 61..., [4] has been deduced previously [3,5] from Coulomb explosion time of flight experiments to be a planar D_{6h} geometry [6], but the key aspect of the present paper is that these first rotational spectroscopy results should provide an unambiguous and precise determination of structure. The geometries of the clusters deduced from the measured rotational constants are consistent with the authors own classical mechanical simulations [7] (which neglected thermal motion of the ions within the clusters) and with the previous Coulomb explosion measurements [3,5]. The clusters appear to be entirely rigid with no observed effects of centrifugal distortion up to $J = 200$. The transitions are believed to be magnetic dipole in origin, the very high orbital angular momentum (with all electrons orbiting synchronously [7]) giving rise to very large magnetic dipole moments. Previous evidence is said to show that no quantized vibrations exist [8, 9, 10, 11] and that there is a more or less continuous phonon distribution. The clusters are metallic in the sense that there is electron delocalisation and a zero band gap. Despite their formation at a temperature of up to 900K, the clusters self cool by a combination of stimulated emission and electron evaporation effects leading to a "kinetic temperature" of 20K [5].

Holmlid and coworkers have also published a number of articles which propose or report: that Rydberg Matter can be used as the active medium in a thermal ultra-broadband tunable laser [12,13]; that the nuclei of comets are covered with an atmosphere of Rydberg species and Rydberg matter [14]; that the diffuse interstellar bands [15] and unidentified infrared bands [16] may be attributed to electronic excitation in Rydberg Matter; that the stable exospheres on the Moon and Mercury are explained by heavy Rydberg Matter clusters [17]; that Rydberg Matter is at least part of the dark matter of the Universe [18] and can be used to explain the Faraday rotation results for the transmission of radiofrequency waves in intergalactic space [19]; that atomic Hydrogen RM clusters are potentially an energy-rich fuel [20, 21]. In one of these papers the authors state "RM is a state of matter of the same status as liquid or solid" [14].

Given the authors' claims to importance of this new state of matter, it is perhaps surprising that there has been very limited quantitative theoretical work on these RM systems, and that most of the experimental work has been confined to one group. The Editors hope that the publishing of these high-resolution spectroscopic results on Rydberg matter clusters will give theoreticians a benchmark to develop accurate quantum mechanical and/or molecular dynamics methods capable of predicting the structure and dynamics of these clusters and their spectroscopic transitions and transition probabilities [22, 23]. This paper may also stimulate experimentalists from other groups to develop measurements in this area. It seems to the Editors that additional work to understand the mechanism for self-cooling of the clusters and the apparent lack of interaction between electronic and vibrational motion (i.e., the motion of the ions) would be highly beneficial. The K_{19} RM clusters observed here, in which each atom is in an $n = 4$ level, have each about 50 eV of internal electronic energy. It is intriguing that

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3 this excess energy does not lead either to fragmentation or to ionization, the narrow lines of
4 the transitions, 20 Hz, suggesting lifetimes of several milliseconds. K_{19} RM clusters thus seem
5 to exhibit a behaviour that is quite distinct from that expected on the basis of current
6 knowledge of intramolecular dynamics in normal gas-phase molecules or clusters. It is
7 suggested in the present paper that these clusters emit microwave radiation by stimulated
8 emission in pure magnetic dipole rotational transitions from high rotational levels. The
9 mechanism proposed for the production of the high rotational levels, via de-excitation from
10 high ℓ to low ℓ states, with rotational excitation of the clusters produced to conserve angular
11 momentum, seems to be rather different from the established behaviour of microwave
12 transitions in isolated Rydberg molecules i.e., that single-photon transitions induce small Δn
13 and $\Delta \ell = \pm 1$ changes and that the rotation of the molecular core remains largely unaffected.
14 The nature of the couplings of angular momenta in these Rydberg matter systems remains to
15 be established, but if the coupling of the electronic and nuclear motion were effectively strong
16 enough for large changes of the rotational motion to take place, then the excitation of cluster
17 vibrations would also seem to be a likely result. Finally, to detect pure rotational magnetic
18 dipole transitions in such systems with the antenna-like detectors used in the experiments
19 reported here we estimate that very high densities of RM clusters must be present in the
20 apparatus. The efficiency of the formation process of the RM clusters, which involves a
21 transition from an initially incoherent cluster of excited atoms into a stable highly correlated
22 electronic state, must therefore be remarkably high. The relationship between this process and
23 the formation of ultracold Rydberg gases and plasmas, in which coherences are created by
24 laser excitation, remains to be clearly established. There are also questions of how these
25 results relate to experimental studies of Rydberg molecule scattering at surfaces [24].
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33 As Editors of Molecular Physics, we would welcome more submissions that shed light on
34 these questions.
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36 T. P. Softley, F. Merkt
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