CHARLOTTE FROESE FISCHER (1929–2024): HER LIFE IN AND CONTRIBUTIONS TO SCIENCE

SUMMARY: Charlotte Froese Fischer's scientific career spanned eight decades that dominated the field of computational atomic physics, radiative processes, and electron-atom collisions.

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Professor Charlotte Froese Fischer might be best known as a pioneer in computational atomic physics, developing methods and methodologies to represent a wide range of atomic properties. Her early work covered electron correlation in atomic systems, but later she extended this to a wide range of topics involving complex interactions with radiation and collision phenomena. The following article first outlines her early scientific career and then enlarges on her many collaborations and her monumental influence on the field.

Charlotte began her education in Vancouver, Canada. She attended the University of British Columbia (UBC) there for her BA degree in Mathematics and Chemistry in 1952 and her MA in Applied Mathematics in 1954. She moved to Cambridge, England, for her Ph.D., supervised by Douglas Hartree, and naturally her paper with him [1] concerned the solution of the Hartree-Fock (HF) equations for states of Ne^{3+} and Ne^{4+}. On her return to UBC in 1957 as a professor in the Department of

Mathematics, she pursued her HF calculations of other complex ions. But in keeping with her mathematical background, she also studied mathematical aspects of these nonlinear integro-differential equations, including their convergence to self-consistency. In 1964, in recognition of her achievements, Charlotte was the first woman to be awarded an Alfred P. Sloan Fellowship.

While at UBC, she had access to a large-scale computer. It was clear that the HF equations had limitations for accuracy, so she developed extensions to the theory to incorporate many configurations. This multi-configuration Hartree-Fock (MCHF) approach [2,3] proved very powerful and thereby results of much greater accuracy were achieved [4,5]. Charlotte introduced the idea of relaxing the radial orbital orthogonality constraints to capture electron correlation more efficiently [3,6]. The standard Racah-Fano algebra for the angular integration of the Hamiltonian had to be extended [7] and the angular codes were adapted accordingly [8]. With increasing computing power, she and her collaborators were able to undertake sequences of calculations which showed the convergence of results as more terms were systematically introduced into the MCHF wave functions [9,10]. This gave much greater confidence in the accuracy of the largest calculations. She was never happy to extrapolate her results to a supposed "best" value, but there was really no need to do that – everyone trusted her results, and with good reason.

In the 1990's Charlotte made major contributions to the understanding of correlation in negative ions [11], including being the first to predict a positive electron affinity for calcium [12] (a discovery that led to her becoming a Fellow of the American Physical Society). The work on negative ions also included important contributions to the prediction and understanding of transitions between bound or semibound states of negative ions [13], a topic still of great importance for example in sympathetic cooling [14]. About the same time, an increased interest in hyperfine structure effects and isotope shifts led to new predictions and exotic discoveries in astrophysics, for example long-lived states decaying only with hyperfine-induced transitions [15,16] and *F*-dependent lifetimes [17]. Similar methods also explained and predicted transitions induced by an external magnetic field [18,19]. During this time, she also engaged in the forefront of computing intercombination lines [20] and *J*-dependent lifetimes [21,22].

Relativistic corrections need to be included at some stage to achieve reliable results with increasing nuclear charge. Charlotte first developed the MCHF + BP method that introduces relativistic corrections in the Breit-Pauli (BP) approximation using the configuration-interaction (CI) approach to improve atomic wave functions [23,24]. In that context, a Davidson program was built [25] to find a few selected extreme eigenpairs of a large, sparse, real, symmetric matrix. This efficient algorithm and code remain a cornerstone of the most recent atomic structure packages for solving the eigenvalue problem targeting the physical atomic energy levels of interest.

In parallel with this, Charlotte and co-workers continued pushing the limits for accuracies in the treatment of correlation [26]. In some cases, this gave results approaching the spectroscopic accuracy of experiments [27], thereby opening the way to tests of quantum electrodynamic (QED) effects in rather complex atomic systems [28]. All this led to a close synergism both with computational science

and technology when developing new methods, but also with experimental physics, fusion research and astrophysics.

Early on, Charlotte engaged herself in parallel computing. In 1992 she presented a paper on a hypercube conversion of serial codes for calculations of atomic structure [29]. A few years later, she and her post-docs presented a Parallel Virtual Machine (PVM) implementation of the MCHF package [30]. These works set the standard, and from this time onwards all her codes were adapted for parallel computing on supercomputers. Dynamic memory allocation, sparse-matrix methods, combined with Gaigalas' angular library [31,32] were implemented in the atomic-structure package ATSP2K [33] to allow large-scale calculations of atomic energy levels, transition rates of all types, isotope shifts, hyperfine constants, and Land\'e factors. For transition probabilities, the individual orbital optimization of wave functions for the initial and final states produces the most accurate wave functions for given expansions but complicates the calculation of the transition matrix elements since the two sets of orbitals will be nonorthogonal. A biorthonormal transformation of the orbitals, with a counter-transformation of the MCHF/CI eigenvectors that leave the total wave functions invariant was implemented to solve this issue [34].

When the MCHF + BP approach alone could not provide the results she wanted, (for instance when the Breit-Pauli approximation becomes inadequate to describe both correlation and relativity), Charlotte was happy to embrace other codes and methods to meet her needs [26]. In particular, she led the team developing Ian Grant's fully relativistic GRASP code [35,36]. That team continues to work together [37,38] through the international Computational Atomic Structures (CompAS) network (see Fig. 1 for a photo of the group at the 2016 Malmö meeting). The latest release of the code, GRASP2018, with Charlotte as the main author [39], has the same broad coverage of properties as ATSP2K. In addition, it comes with a detailed manual [40], allowing a broad range of researchers to compute atomic data of importance in various fields such as astrophysics, nuclear physics, and medicine. Examples of applications, to mention a few, include computation of atomic data needed for the extraction of nuclear radii from laser measurements on isotope separators [41,42], along with transition and energy data for astrophysical element abundance determinations [43-46]. It is not an overstatement to say that the code is a very important working horse in many fields.

Along with her calculations, Charlotte was very committed to making her computer programs available and was one of the first subject editors of the journal *Computer Physics Communications*. This ensured that those who developed such programs were given due acknowledgement of their work. She contributed her own codes and worked with other authors to enhance their codes to make them even more applicable than the authors had originally intended.

In addition to her remarkable work on electronic structure discussed above, it was unavoidable for Charlotte to move beyond pure atomic structure and include the treatment of continuum states --- first with auto-ionization [47,48] and photo-ionization [49,50] and later with collisions. This also led to further development of the spline Galerkin method [51], both for continuum processes and bound states. Charlotte's work with the late Oleg Zatsarinny had a tremendous impact on the calculation of electron collisions, especially from complex targets. A very important paper to introduce what later

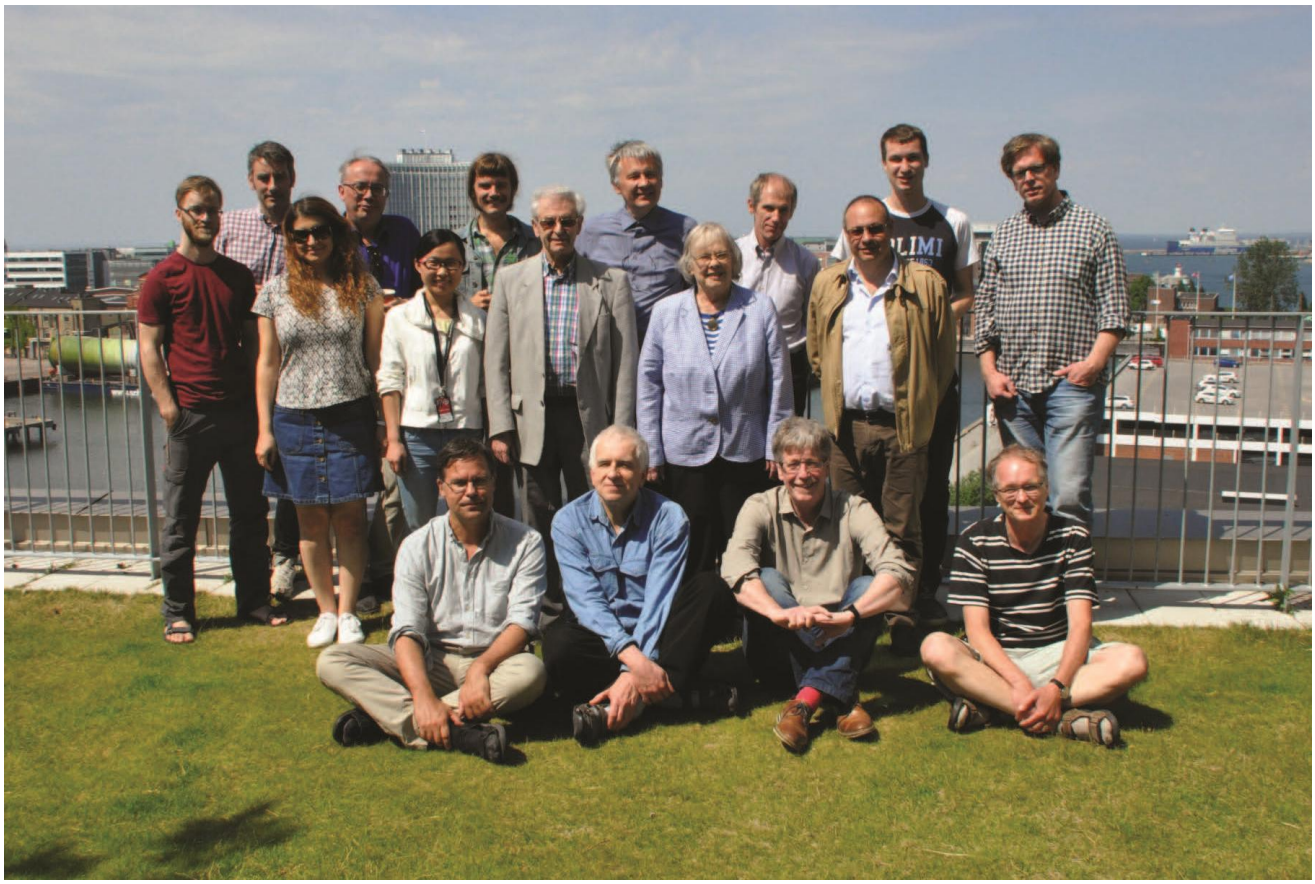

Figure 1. Group picture taken at the Malmö CompAS meeting, June 2016. Charlotte is standing in the middle row, second from the right.

became known as the B-spline R-matrix (BSR) method was the one on photo-ionisation of lithium [52]. Recall that photo-ionisation is essentially electron scattering from the residual ion; *i.e.*, one needs to solve that problem and generate matrix elements with the initial bound state. Charlotte had generously invited Oleg to Vanderbilt University after science in the Ukraine became difficult, and she also shared her B-spline library with Oleg to adapt for his purposes. This ultimately resulted in Oleg's most important contribution to science, namely the publication of his computer code in CPC [53]. Charlotte and Oleg kept working together as an excellent team for many years, with her mostly being interested in numerical methods and their application to structure physics, while Oleg's emphasis was on electron collisions. Being a general close-coupling code, BSR can be run in complete bound-state mode (for a comparison with MCHF, see [54]), for photo-ionisation as a bound-continuum process, and for electron collisions. To get an impression of what can be done with BSR, see the Topical Review [55]. The extension from a non-relativistic and semi-relativistic (Breit-Pauli) to a full-relativistic (Dirac-Breit) version, once again, critically depended on Charlotte's collaboration [56]. The importance of BSR is seen in its prominent status as a contributing software to fill many databases, including LXCat [57].

Without Charlotte's inviting Oleg, this work would most likely never have reached the current status. In 2021 Charlotte published a review of these developments [58].

A somewhat different aspect of Charlotte's influence flowed from a panel discussion on uncertainty quantification held as part of the 2010 ICAMDATA meeting in Vilnius, Lithuania. The participants were Charlotte Froese Fischer and Igor Bray, with Gordon Drake as moderator. The purpose of the discussion was to ascertain the degree of support from the theoretical atomic physics community for a new policy to require uncertainty estimates for computations of atomic properties. Charlotte's support played a key role in establishing the new policy, and it is now widely regarded as the standard for publications in the field.

Her books [3,59] have been very influential for new generations of atomic physicists. An important autobiographical article entitled "Reminiscences at the end of the Century" [60] appeared in a special issue of *Molecular Physics* that commemorated Charlotte's scientific achievements and celebrated her 70th birthday. Further details can be found in the review article [61] that appeared on the occasion of her 90th birthday.

Charlotte's legacy is, of course, the quality of her work, but it also includes the supportive way she worked with colleagues and students alike. She will remain an exemplary educator and a role model for us all.

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