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DEPARTMENT OF CHEMISTRY DAVIS, CALIFORNIA 95616

February 8, 2022

Prof. Petr Slavíček, Chairman,
Habilitation Committee
Department of Physical Chemistry,
University of Chemistry and Technology, Prague
Technicka 5
Prague 6, 166 28
Czech Republic

Dear Prof Slavicek,

I am writing in strong and enthusiastic support for the promotion of Dr. Přemysl Kolorenč to the rank of Associate Professor and approval of his habilitation thesis. To establish my perspective on his work, let me say that my own research in the theory of electron-molecule collisions, molecular photoionization, ultrafast and multiphoton spectroscopy over the last 35 years has frequently made use of the developments during that time in the groups of Professors Cederbaum and Domcke that form many of the underpinnings of the work on interatomic Coulombic decay that is described in Dr. Kolorenč's thesis.

Informed by my familiarity with that literature, I can say with confidence that Dr. Kolorenč's thesis is a *tour de force* description of a comprehensive body of work in this area. Interatomic Coulombic decay was predicted by Professor Lenz Cederbaum in the late 1990s, and since then it has become a focus of both experimental and theoretical research in atomic and molecular physics. The primary reason for this interest is that, despite its recent discovery, this phenomenon is evidently a nearly universal feature of the response of matter to ionization that removes inner valence or core electrons from an atom or molecule. X-ray spectroscopy at synchrotron and other light source facilities around the world has been routinely able to access this sort of ionization for decades. Nonetheless, it was the only the application of coincidence measurements (in particular the cold target ion recoil momentum spectroscopy or COLTRIMS method) that revealed unambiguously the phenomenon of ICD and its mechanism.

The first of those experiments were performed while Dr. Kolorenč was a Ph.D. student at Charles University. There he worked with Professor Jiří Horáček on topics in electron-molecule collisions and metastable anions and clearly mastered the techniques of theoretical collision physics. The remainder of his publication list reads like a history of the development of the theory of interatomic Coulombic decay as well as some of the seminal experimental work on the phenomenon. It is not possible to study this field without repeatedly encountering the work of Dr. Kolorenč.

Immediate evidence of the impact of the work described in this thesis can be seen in Dr. Kolorenč 's collaboration with the group of Horst Schmidt-Böcking and Reinhart Dörner in Frankfurt, which developed COLTRIMS coincidence measurements. It is clear in the beautiful work in 2010 with Dr. Till Jahnke in Franfurt described in publication A9 that the theoretical work of Dr. Dr. Kolorenč and his collaborators guided the conception of these experiments. In that experiment one helium atom in the He_2 dimer is simultaneously ionized and excited to its n=2 states. The He_2 dimer is the most quantum

mechanical of all diatomic molecules since its almost vanishingly small potential well binds only one vibrational state that spends about 80% of its time in the tunneling region. That state extends out to an internuclear distance of around 100 Å. Its motion after the initial ionization as the molecule contracts and undergoes interatomic Coulombic decay is written in the momenta of the He^+ atomic fragments that explode apart after that decay. That work is described in the section of the thesis that treats " Ultralong-range energy transfer via ICD," and in the set of publications A9-A11. It is landmark work, breathtaking in the detailed and quantitative theoretical description of this experiment that it provides.

Let me turn now to the fundamental theoretical problem that this habilitation thesis treats. Interatomic Coulombic decay, like Auger decay, is purely a phenomenon of electron correlation. In quantum chemistry electron correlation is the correction to the Hartree-Fock or mean field theory that generally gives more than 95% of the energy of molecules and adequately describes chemical bonding. In the problem treated by this thesis electron correlation, the central challenge of quantum chemistry, is not a correction. It is everything. Electron correlation is the mechanism by which these decays occur. For that reason, the thesis focuses attention on the work in paper A1 and the contributions of the candidate to the quantum chemistry method known as algebraic diagrammatic construction (ADC).

The signature accomplishment of Dr. Kolorenč in this area is the development of critical components of the technique that is known as the Fano-ADC method. The ADC method is a computational electronic structure method developed primarily in the Cederbaum group in Heidelberg. To apply it to the calculation of the lifetimes of metastable states that decay via Auger or interatomic Coulomb decay, ADC must be combined with the technique of Stieltjes imaging to extract that information from the finite basis set calculations of quantum chemistry. That clever combination of ideas, reaching back to the work of Andrew Hazi and Peter Langhoff in the 1970s, opened the door to applying this idea of bound state quantum chemistry to autoionizing metastable states. The ADC(2,2) approximation that adds the class of three-hole two-particle excitations to this method in its application to autoionizing states was a necessary step towards making this approach capable of reliably quantitative predictions. I think that development is all Dr. Kolorenč's work.

The thesis is clearly and pedagogically written. In its section on "Extracting R-dependent absolute decay widths from the experiment" the shortcomings of the theoretical methods in this thesis (and all others for that matter) for computing decay widths are forthrightly described. The section on "Outlook" succinctly summarizes both the opportunities and challenges that will animate Dr. Kolorenč's work in the immediate future. All of the components are in place to ensure a productive and impactful future career for him.

I urge you, on the basis of his habilitation thesis and his published track record of extraordinary research, to approve this thesis and to promote Dr. Přemysl Kolorenč to the rank of associate professor. I would be thrilled to have him as a colleague, and I think you should be too.

Sincerely,

C. William McCurdy
Distinguished Professor of Chemistry
University of California, Davis
and
Associated Faculty at the Lawrence Berkeley National Laboratory

P.S. I have gone through the check of originality of the thesis done by the system Turnitin and it is clear to me that the thesis represents an original work with the only meaningful overlap with prior work being with the existing literature written by the author. In my opinion, the (auto)plagiarism audit (Turnitin report, in Czech) did not indicate scientific misconduct regarding copying.

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