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Effects of static electric fields on the photoionization spectra of two-electron atoms and ions

Monica Halka*

Embry-Riddle Aeronautical University, Prescott, AZ 86301, USA

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Abstract

The simultaneous photoexcitation of two electrons is a highly correlated process, producing doubly excited states, observed as resonant structure in the ionization cross-section. This review paper explores the ability of a static electric field to affect these correlations, as evidenced by changes in the shape and energy of resonances. A comparison of field effects on the spectra of H^-, Ps^-, He, Ba and Cs^- summarizes current understanding of the processes and the capability of theory to predict the cross-sectional features.

Keywords: Autoionization; Barium; Correlation; Doubly excited states; Electric field; Helium; Negative ions; Photoionization; Resonances

1. Background

In 1978, first experiments reported the effect of applying a static electric field to the interaction region for photodetachment of H^- near the threshold for production of neutral hydrogen in the $n = 2$ state (Gram et al., 1978). This study, combined with later experiments on higher-n threshold regions (Halka et al., 1993), showed that the doubly excited states of H^- are particularly sensitive to electric field strength, but in some unexpected ways.

Doubly excited states were discovered in the late 1950s in inelastic scattering of electrons by helium (Silverman and Lassettre, 1958). Since that time these so-called Feshbach or 'closed-shell' resonances have been observed in photoexcitation spectra of the negative ions of hydrogen, sodium, and chlorine as well as in the spectra of neutral atoms like lithium and barium. In general, these states are autoionizing, appearing above the energy threshold for detachment of a single electron and therefore in the continuum of free electrons.[1] The autoionization process, where one electron escapes and the other ends up in a lower-n state, is necessarily continuum-assisted.[2]

These states may be seen as resonances by considering a rather simple view of the two electrons on opposite sides vibrating simultaneously towards and away from the nucleus. This electron-correlation behavior has been dubbed the '+' mode and is responsible for most of the observed cross-sectional structure. (The '−' mode occurs when the electrons vibrate in phase, producing a much weaker resonance effect.) Correlation is strongest when

[1]Barium is a noteworthy exception, having doubly excited bound states.

[2]The term 'autoionization' is only strictly valid when one is discussing doubly excited states of neutral atoms. For doubly excited states of negative ions, 'autodetachment' is more generally accepted.

*Tel.: +1 928 777 3876; fax: +1 928 777 3827.
E-mail address: monica.halka@erau.edu.

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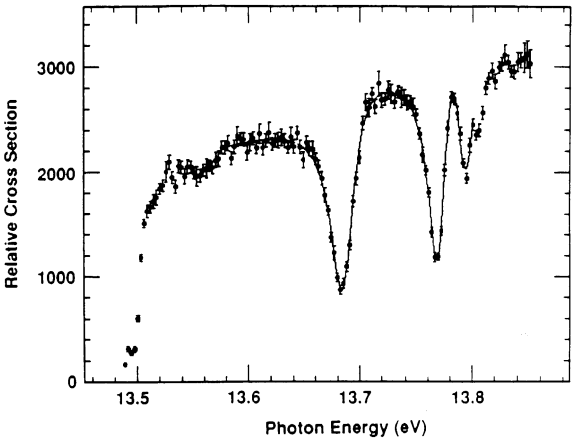

Fig. 1. Photodetachment of H^- with detection of fragment H(4) atom. Applied field, $F = 0$ kV/cm (Harris et al., 1990).

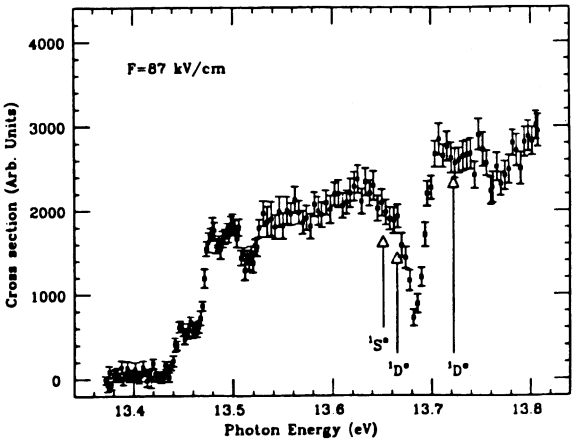

Fig. 2. Photodetachment of H^- with detection of fragment H(4) atom. $F = 87$ kV/cm. Arrows point to field-free central energies of some even parity resonances (Halka et al., 1993).

the two outer electrons are initially excited to the same principal quantum number. Resonances are also observed where the electrons are excited to n and $n+1$, n and $n+2$, n and $n+3$, etc. Thus, a series is usually observed converging to the energy threshold for production of a residual atom or ion in the $n+1$ state. Fig. 1 shows such a series of doubly excited '+' states in H^- converging to the $n=5$ production threshold (13.8 eV) of neutral hydrogen. Note that these are all $^1P^o$ states, owing to angular momentum selection rules, and appear as dips or 'windows' in the cross-section, rather than peaks. This is not the case in general—the form depends on varying core effects in different atomic species and is reflected in the asymmetry parameter of the Fano profile (Starace, 1977).

2. Field effects

Fig. 2 shows the same region of the photoexcitation spectrum, but with an applied field of 87 kV/cm. The most obvious change is in the threshold region. In the zero-field case, doubly excited $n=5$ states can decay only to neutral hydrogen with $n \leqslant 4$. So members of the series approaching the 13.5 eV threshold from below would not appear in this spectrum. With an applied field, however, one electron is still ejected, but *the other need not decay to a lower n-state.* The field supplies the energy needed to release the outer electron. Consequently, we are able to observe resonances decaying to the 'parent state.' The threshold shift itself has been calculated by Zhou and Lin (1992), who find that a weak residual coupling allows the states associated with the '+' channel to decay through the '−' channel, resulting in a shift close to the measured value.

Now let us look at what is happening to the series approaching 13.8 eV. At first consideration, it seems that most of our series disappears or is 'quenched' by the field, while the strongest resonance suffers a shift in energy. Its shape also appears to change. This may be attributed to the emergence of neighboring states with $\ell \neq 1$, which is allowed when a field is present to provide mixing. In this case, the even-parity states with $\ell = 0$ or 2 are distorting the $^1P^o$ profile.

A clearer picture of Stark splitting is seen in the spectrum of the $n=2$ Feshbach resonance subjected to a static electric field of 110 kV/cm (Fig. 3). Note that the $^1P^o$ resonance shifts about 5 meV lower in energy. Theoretical work using stabilization (Callaway and Rau, 1978) and complex coordinate methods (Wendoloski and Reinhardt, 1978; Ho, 1995) were successful in describing the overall behavior. Such resonance-energy shifts due to spectral repulsion among Stark states in changing applied fields are also theoretically predicted for the very similar case of Ps^- (Ho and Ivanov, 2000).

So we see that resonances are not quenched by the field, as previously assumed. That is, until the field is of sufficient strength, it does not detach the outer electron, but redistributes the resonant structure to other allowable Stark states in a rather complex manner. This has recently been beautifully demonstrated in experimental work of Bates et al. (2001). Fig. 4 shows their photoexcitation spectrum of barium with varying applied field strengths, where energy sharing among states is evident. Though the configuration interaction is strong, the authors find closed-orbit theory adequate to describe the spectral structure.

Recent work on the photoionization of helium in the presence of an electric field reveals a complex redistribution of states (Fig. 5). Though theory (Chung et al., 2001) did not predict the abundance of new structure at