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USING NUCLEAR RESONANCE EXCITATION TO OBSERVE UNITED ATOMS IN SYMMETRIC ION-ATOM COLLISIONS

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Summary

We consider nuclear resonance excitation as a means to observe the distribution of united-atom orbitals in symmetric ion-atom collisions. We develop this possibility with the application of a two-state adiabatic model to an analysis of a ^{8}Be nuclear resonance experiment. To test our model, and to provide a more direct determination of the nuclear resonance width, we propose that the ^{8}Be experiment be repeated with metastable helium targets.

Introduction

The old notion of "sticky" nuclei as tools for studying atomic inner-shell vacancy production has come to be fully appreciated only recently, and mostly as a result of the innovations of John Blair and his coworkers in Seattle on experiments with light nuclei as targets, and protons as projectiles.

Almost twenty years ago, Ciocchetti and Molinari[1] proposed that inner-shell excitation be observed in the region of a nuclear resonance whose lifetime is comparable to inner-shell periods. They showed that the excitation could be measurably affected by the nuclear time delay imposed on the electron-projectile interaction[2]. Although the Ciocchetti-Molinari model accounted for the sticky nuclei in a reasonable way, the nuclear time delay proved to be too awkward to evaluate, and therefore, to be useful quantitatively.

Fifteen years later, Blair revived[3] the Ciocchetti-Molinari model by demonstrating that the nuclear time delay could be replaced by a nuclear resonance phase shift, thus allowing the notion of sticky nuclei to become operational. In a time-independent energy representation, he showed that, when the width of resonance is comparable to the electron ionization energy, the nuclear time delay manifests itself in the excitation as an interference between two nuclear resonance amplitudes evaluated at different energies, since a projectile which ionizes an electron on the way "in" reaches the nucleus with a lower energy than one which ionizes on the way "out".

Petitioning Blair's idea, the Seattle group bombarded ^{58}Ni with resonant protons. They observed that the coincidence rate for creating a K-shell vacancy with the scattered proton displays the predicted dependence on the incident energy[3]. The Blair model has been subsequently established theoretically through independent studies[4], as well as experimentally on a variety of systems[5].

With this talk, we shall extend the notion of sticky nuclei as probes of inner-shell excitation to symmetric, or nearly symmetric, ion-atom collisions. We shall consider resonant systems in which both the collision time and the nuclear time delay are comparable to electron orbital periods. Such collisions are properly described as the evolution of quasimolecular orbitals (MO); as the internuclear separation changes with time, electrons are promoted (or demoted) to other MO at the expense (or gain) of energy of the nuclear reaction partners.

The Blair model, based on the semiclassical excitation amplitudes for asymmetric ion-atom collisions, fails to recognize the MO promotion mechanisms inherent in more symmetric collisions.

To discern these mechanisms fully, we shall adapt a two-state MO model with the introduction of a resonant phase shift to represent the courtship of the sticky nuclei. Since this resonant relationship examines the molecular orbitals at an unconventional point, the united-atom apex of their journey from one separated-atom limit to another, experiments of the type we will consider could provide new tests of MO promotion models.

The ^{8}Be Resonance Experiment

Over twenty years ago Hans Staub and coworkers in Zurich initiated a precision experiment to determine the width of a narrow ^{8}Be nuclear resonance[6]. It wasn't until four years later, after having achieved an instrumental resolution of 95 eV (remarkable, at 200 keV, even by today's standards), that they bombarded ^{4}He with 184 keV (Lab) $^4He^+$ and determined the width, $\Gamma \simeq 13.6$ eV (Lab), of the ^{8}Be nuclear ground state[7,8]. A fascinating account of this experiment has been provided by Staub and his group in "Adventures in Experimental Physics".[9]

At 200 keV, the He^+ ion enters the helium atom with a speed almost equal to that of a helium electron. The collision is certainly not adiabatic, but nevertheless, the atomic electrons evolve via MO into states of the united-atom ion Be^+. Since the width of resonance is small compared to the Be^+ electron-level separations, $\varepsilon^{UA}_{2p} - \varepsilon^{UA}_{1s} \simeq 120$ eV, the Zurich group observed a splitting of the nuclear excitation function into a series of structures representing the occupation of the united-atom levels during the nuclear resonance.

Their data has been reproduced in Fig. 1. The curve through the data represents their best numerical fit, after folding in the instrumental resolution, to determine, among the relative intensities of the atomic structures, the width and position of resonance.

It is intriguing that, unlike the experiment of Blair and coworkers, the Zurich experiment was not a coincidence observation, since no determination of the state of the atomic electrons was made other than that inferred from the fragmentation of the nuclear excitation function. It is the presence of the sticky nuclei which provides a close look at the distribution of the molecular orbitals in the united-atom limit.

Two-State Molecular Orbital Model

To present our ideas clearly, we restrict ourselves to the elastic channel, in which the target and projectile begin and end the reaction in their ground states, and retain a simple two-state approximation ignoring all couplings among the MO[10]. We will single out an active 1s electron and refer to the $1s\sigma_g$ and $2p\sigma_u$ MO, $X_g(\vec{r},R)$ and $X_u(\vec{r},R)$, with eigenenergies $\varepsilon_g(R)$ and $\varepsilon_u(R)$, respectively.

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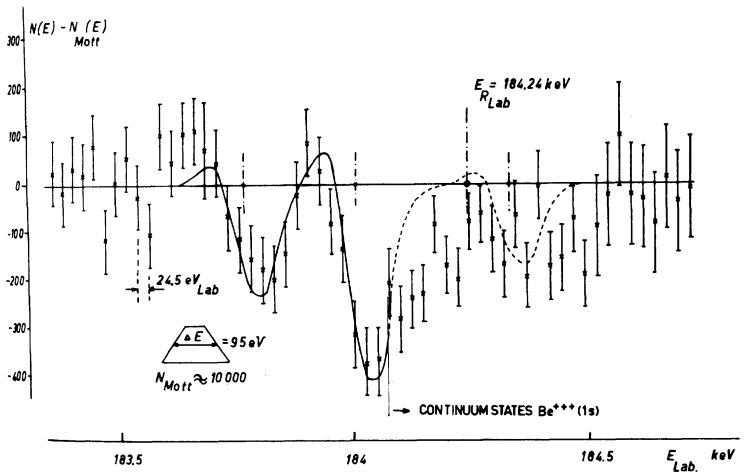

Fig. 1 ^{8}Be excitation yield, actual counts minus the symmetrized-Coulomb (Mott) background (see eq.(10)), as a function of the incident He^+ energy, reproduced from ref. 8. The first, low-energy dip corresponds to the $Be^+(1s^22p)$ united atom.

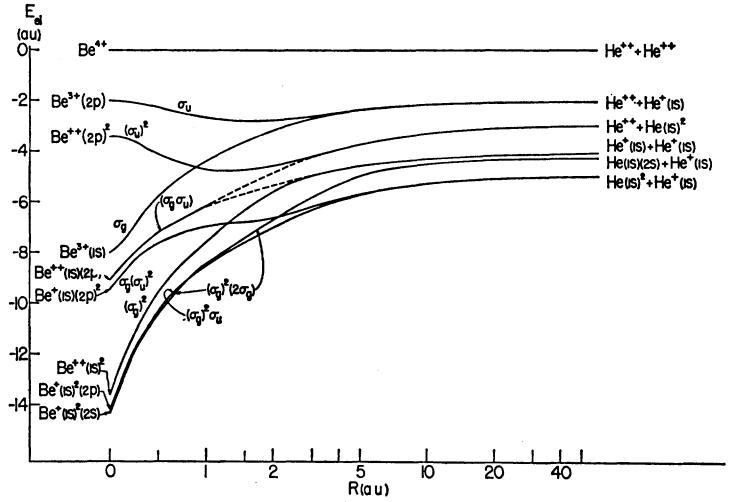

Fig. 2 Diabatic curves suitable for a discussion of non-adiabatic collisions involving He ions, reproduced from ref. 11. Here $E_{el} = \varepsilon_\nu(R)$ from the text, and one atomic unit (au) of energy equals 27.2 eV.

Here R is the internuclear separation, and $\vec{r}$ is the position of the electron relative to the center of mass of the nuclei. The indices g and u label, respectively, states symmetric and antisymmetric under interchange of the identical nuclei.

Our discussion can be generalized somewhat by consideration of diabatic three-electron states, $(1s\sigma_g)^2(2p\sigma_u)$ and $(1s\sigma_g)(2p\sigma_u)^2$, whose eigenenergies as a function of R have been reproduced in Fig. 2. As discussed in detail by Lichten[11], the diabatic states are more appropriate for the study of nonadiabatic collisions of interest to us here.

It is surprising that even the elastic channel contributes to the fragmentation of the nuclear excitation function. An examination of the energy levels in Fig. 2 reveals that the initial system state, $He(1s^2) + He^+(1s)$, evolves as $R \to 0$ via the $1s\sigma_g$ and $2p\sigma_u$ orbitals into two united-atom states, $Be^+(1s^22p)$ and $Be^+(1s2p^2)$, in which the nuclear resonance occurs. The Coulomb binding of the electron ensures that the system's energy is conserved, and, consequently, that the resonance energy is effectively lowered in the $1s\sigma$ channel and raised in the $2p\sigma$ channel.

Initially, for large R, we assume that the electron is bound to the target nucleus 2 in the ground state $\psi_{1s}(\vec{r}_2)$ with energy ε_{1s}. At infinite separation the molecular wavefunctions, X_g and X_u, correspond to the same energy ε_{1s} and

$$\begin{aligned} \tfrac{1}{2}(X_g + X_u) &\sim \psi_{1s}(\vec{r}_2) \\ \tfrac{1}{2}(X_g - X_u) &\sim \phi_{1s}(\vec{r}_1) \end{aligned} \quad \text{as } R \to \infty, \tag{1}$$

where $\phi_{1s}(\vec{r}_1)$ is the charge-transfer state of the electron bound to the projectile 1.

We expand the solution of the system wave equation in terms of the two states, $1s\sigma_g$ and $2p\sigma_u$, and ignore all other molecular orbitals:

$$\Psi = F_g(\vec{R})\, X_g(\vec{r},R) + F_u(\vec{R})\, X_u(\vec{r},R). \tag{2}$$

The expansion coefficients, $F_\nu(\vec{R})$ (ν = g or u), are interpreted as the wavefunctions of relative nuclear motion and define scattering amplitudes in the large R limit by

$$F_\nu \underset{R\to\infty}{\sim} \tfrac{1}{2}\left(e^{i\vec{k}\cdot\vec{R}} + f_\nu(\theta)\,\frac{e^{ikR}}{R}\right) \tag{3}$$

where θ is the scattering angle and k is the wave number determined by the system's initial energy,

$$E = \hbar^2k^2/2\mu_{12} + \varepsilon_{1s} \equiv K + \varepsilon_{1s}.$$

Consequently, the system wavefunction can be written asymptotically as

$$\Psi \underset{R\to\infty}{\sim} e^{i\vec{k}\cdot\vec{R}}\,\psi_{1s}(\vec{r}_2) + \frac{1}{2}\frac{e^{ikR}}{R}\,[\phi_{1s}(f_g-f_u) + \psi_{1s}(f_g+f_u)], \tag{4}$$

so that the differential cross section for elastic scattering in the system center of mass is given by

$$\frac{d\sigma_{elas}}{d\Omega} = \frac{1}{4}\,|f_g+f_u|^2. \tag{5}$$

The nuclear wavefunctions satisfy in the adiabatic limit the decoupled equations

$$[-\frac{\hbar^2\nabla^2_{\vec{R}}}{2\mu_{12}} + V(\vec{R}) + E - \varepsilon_\nu(R)]\, F_\nu(\vec{R}) = 0 \qquad (6)$$

where $V(\vec{R})$ is the internuclear (Coulomb plus strong) potential, and ε_ν is the MO eigenvalue.

The colliding nuclei essentially follow a Coulomb trajectory, and the electron, sharing the system's energy, serves only to accelerate (or decelerate) the nuclear motion along that trajectory. In this spirit, and utilizing the short range on atomic scale of the stronger nuclear interaction, we approximate eg. (6) by replacing the molecular eigenvalue with its united-atom limit $\varepsilon_\nu(R\to 0) \equiv \varepsilon_\nu^{UA}$. We infer that the system resonance energies are a superposition of the united-atom spectrum with the nuclear spectrum K_n:

$$E_{n_\nu} = K_n + \varepsilon_\nu^{UA}$$

In the case of the ^{8}Be ground state resonance, the nuclear scattering can be represented by a single s-wave resonant phase shift along with the Coulomb scattering amplitude[8]:

$$f_\nu(\theta) = f_\nu^{C}(\theta) + f_\nu^{R}(\theta), \qquad (7)$$

where

$$f_\nu^{C}(\theta) = -\frac{\eta}{2k}\,\frac{\exp[-2i\eta\ln\sin(\theta/2)]}{\sin^2(\theta/2)},$$

$$f_u^{C}(\theta) = f_g^{C}(\pi-\theta), \qquad (8)$$

and

$$f_\nu^{R}(\theta) = \frac{1}{2ik}(e^{2i\delta_\nu} - 1)$$

$$= \frac{1}{k}\,\frac{\frac{1}{2}\Gamma}{E-E_{o\nu}+\frac{1}{2}i\Gamma}. \qquad (9)$$

Since the Be^+ atomic lifetimes are much longer than the nuclear resonance lifetime, the width Γ reflects that of the nuclear resonance and is independent of the molecular channel.

Here the electron promotion simply shifts the resonance position. This displacement is revealed if we rewrite the energy denominator in eq. (9) in terms of the energy K of relative nuclear motion as

$$E - E_{o\nu} + \tfrac{1}{2}i\Gamma = K - K_o - \varepsilon_\nu^{UA} + \varepsilon_{1s} + \tfrac{1}{2}i\Gamma.$$

With these results, we find that the elastic cross section, eq. (5), can be written as

$$\frac{d\sigma_{elas}}{d\Omega} = \frac{1}{4k^2}\left|\frac{\eta}{2}\left(\frac{e^{-2i\eta\ln\sin\frac{1}{2}\theta}}{\sin^2\frac{1}{2}\theta} + \frac{e^{-2i\eta\ln\cos\frac{1}{2}\theta}}{\cos^2\frac{1}{2}\theta}\right)\right.$$

$$\left. + \frac{\frac{1}{2}\Gamma}{K-K_o-\varepsilon_{1s}^{UA}+\varepsilon_{1s}+\frac{1}{2}i\Gamma} + \frac{\frac{1}{2}\Gamma}{K-K_o-\varepsilon_{2p}^{UA}+\varepsilon_{1s}+\frac{1}{2}i\Gamma}\right|^2. \qquad (10)$$

Comparison with Experiment

Our result, eq. (10), for the elastic cross section is almost identical to that used by the Zurich group in their numerical analysis of the ^{8}Be data. Our adiabatic model determines the strengths of the 1s and 2p united-atom contributions to the resonance splitting to be in the ratio 1:1. The best fit to the data, however, determines this ratio as 3:5[8]. This result is already apparent in Fig. 1, where it is also evident that higher united-atom orbitals have been occupied.

It should not be surprising if our adiabatic model should fail to predict this ratio correctly. The non-adiabatic nature of the collision leads to couplings between the MO. Thus, there could occur a transfer of amplitude from the initial state into the inelastic channels, and consequently, a change of the ratio from its adiabatic value. Ultimately, of course, we would like to evaluate the cross section in a coupled-channels approach including several diabatic states from those represented in Fig. 2. We remark in passing that the potential curves in Fig. 2 were constructed by interpolation between the known separated and united-atom limits; state of the art calculations for $He^+ + He$ have been mostly limited to the $\lesssim$ 10 keV energy range.

In their analysis of their ^{8}Be experiment, the Zurich group assumed that the third electron could be ignored in the united-atom limit, where it is weakly bound (by about 18 eV in the lowest Be^+ state). Thus, they determined the fragmentation of the excitation function by the relative separation of two-electron states: Be^{++} ($1s^2$), Be^{++} (1s2p), etc. Although the three-electron states of Be^+ are probably more appropriate, it is clear, by comparing the united-atom limits in Fig. 2 with the data in Fig. 1, that these two sets of states are indistinguishable. (The difference in their centroids is unobservable, since the nuclear resonance energy K_o is not known absolutely.)

Prediction

In regard to the inelastic channels, it is productive to speculate on the outcome of the ^{8}Be experiment if metastable helium targets were used. We infer from Fig. 2 that the state of the system would evolve as

$$He^+(1s) + He(1s2s) \to (1s\sigma_g)^2\, 2s\sigma_g \to Be^+(1s^2 2s).$$

That is, in sharp contrast to ground-state helium targets, there is a unique united-atom limit in the elastic channel. Furthermore, the rather large separation of the $(1s\sigma_g)^2(2s\sigma_g)$ level from higher levels in Fig. 2 precludes promotion into the inelastic channels. We conclude, therefore, that if metastable targets were used, the nuclear excitation function would not fragment; instead, a single resonance dip would be observed. This experiment would vindicate the MO approach to studies of symmetric collisions with sticky nuclei, and as well, would provide a cleaner determination of the ^{8}Be nuclear resonance width.

The remarkable way in which the ^{8}Be nuclear resonance resolves the united-atom levels is due to the narrow width of resonance compared to the level spacing. Similar resonances exist in other systems. One suitable candidate would be $^4He^+ + {}^6Li$, which has a narrow ^{10}B resonance, $\Gamma \simeq 2.5$ eV (CM), at 448 keV (Lab) incident energy.

In systems with more electrons, the united-atom level separations would generally be comparable to the width of resonance, and there would be interference among overlaping amplitudes from the various united-atom channels. A clear fragmentation of the excitation function, as in the ^{8}Be experiment, would not occur. Nevertheless, careful fits to good data would determine the "spectroscopy" of the united-atom distribution of MO.

Outlook

We have outlined a fully quantum mechanical theory of symmetric ion-atom collisions with sticky nuclei. As in the Blair model of asymmetric collisions, we have exploited the short range of the nuclear interaction on atomic scale to separate the electronic and nuclear excitation amplitudes. Although we have offered a footing for the mechanisms involved, we are several steps short of having developed a practical theory.

We envision a formalism which combines the sticky nucleus amplitude with semiclassical MO transition amplitudes evaluated at the united atom limit. (An R-matrix method which may help achieve this goal has al-

ready been developed[12].) With valid semiclassical MO calculations, at the faster impact velocities which foster sticky nuclei, such a theory of symmetric collisions would be the true operational analogue of the Blair model for asymmetric collisions.

In any event, we have seen that sticky nuclei could provide a powerful probe of symmetric collisions, particularly in the united-atom region, where MO occupations are virtually unobservable by more conventional methods.

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