Approximation Scheme for Electron Capture into Arbitrary Principal Shells of Energetic Bare Projectiles

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An expression given by Fock for the density matrix in momentum space of an electron in the nth hydrogenic shell is used to derive a prescription for calculating capture cross sections into arbitrary principal shells of energetic bare projectiles. Application to an eikonal treatment yields a simple formula showing that capture cross sections scale to the Oppenheimer-Brinkman-Kramers values with a Z-independent factor between 0.1 and 0.4, in agreement with experimental data.

In this Letter we derive a simple prescription which drastically simplifies the calculation of total sections for electron capture[1-3] into arbitrary principal shells of bare projectiles. Such processes become important when heavy, highly charged ions react with atomic hydrogen:

$$A^{+Z}+\mathrm{H}\rightarrow A^{+Z-1}+\mathrm{H}^{+}. \tag{1}$$

This reaction is of technological relevance for tokamak fusion plasmas heated by a neutral hydrogen beam.[4]

While calculations based on classical mechanics[5] have been surprisingly successful in reproducing experimental electron-loss data in the energy range 50–5000 keV/amu most approaches to electron capture using quantum mechanics suffer from the formidable difficulties of handling hydrogenic wave functions with high quantum numbers n. A striking exception in simplicity is provided by the Oppenheimer-Brinkman-Kramers (OBK) approximation.[1,2,6,7] In this particular case, a sum rule first given by Fock[8] and later rediscovered by May[6] allows one to carry out the subshell summations and to derive a closed-form expression. It is well known[9] that the OBK approximation considerably overestimates the experimental total cross section but otherwise reflects the correct behavior. It therefore has become a common practice[1,9] to scale the OBK cross section down by an empirical Z-independent factor $\alpha(v)=0.1$–0.4. So far it is not fully understood why this scaling procedure works so well.

In this Letter we attempt to furnish a theoretical explanation. Our point of departure is the observation that the basic simplicity (regarding states of arbitrary n) of the OBK approximation can be carried over to a more general class of quantum treatments. Let

$$\tilde{\varphi}_{nlm}(\vec{q})=\int d^3r\,\varphi_{nlm}(\vec{r})\exp(i\vec{q}\cdot\vec{r}) \tag{2}$$

be the Fourier transform of the normalized hydrogenic wave function $\varphi_{nlm}(\vec{r})$ and

$$\rho_n(\vec{q},\vec{q}\,')=\sum_{l=0}^{n-1}\sum_{m=-l}^{l}\tilde{\varphi}_{nlm}^{*}(\vec{q})\tilde{\varphi}_{nlm}(\vec{q}\,') \tag{3}$$

be the density matrix in momentum space of the nth principal shell. With these definitions, as early as 1935, Fock[8] has derived the following theorem (atomic units are used throughout):

$$\rho_n(\vec{q},\vec{q}\,')=\frac{2^6\pi n^2 q_n^{\,5}}{(q^2+q_n^{\,2})^2(q'^2+q_n^{\,2})^2}\,\frac{\sin nx}{n\sin x}, \tag{4a}$$

with $q_n=Z/n$ and x denoting a distance on a four-dimensional sphere. In May's paper[6] an expression for the sum of Eq. (3) is given but is left in an undeveloped form. Equation (4a) has also been used by Omidvar[10] in his Born treatment of electron capture into states with asymptotically high n. The physical meaning of the quantity x becomes more transparent by rewriting Fock's expression as

$$\sin\tfrac{1}{2}x=\pm\frac{q_n|\vec{q}-\vec{q}\,'|}{\{(q^2+q_n^{\,2})(q'^2+q_n^{\,2})\}^{1/2}}. \tag{4b}$$

In this form Theorem (4) should be useful for a variety of applications.

It is instructive to first consider the classical limit. The function $d_n(x)=\sin nx/(n\sin x)$ has the value $d_n(0)=1$ and for great n has a narrow peak (of a width $\propto 1/n$) at $x=0$. The backward peak with $d_n(\pi)=(-1)^{n+1}$ disappears if averaged over adjacent n. Hence, for great n, the n-averaged density matrix becomes diagonal as is required for a single classical orbit and hence for the set of orbits with the same energy. The prefactor of $d_n(x)$ in Eq. (4a) is recognized as the classical momentum distribution of a microcanonical ensemble of electrons[1] with energy $E_n=-\frac{1}{2}q_n^{\,2}$ normalized in accordance with Eq. (2) and summed over the n^2 substates.

Rather than taking the classical limit we use Theorem (4) for a simplified *quantal* calculation of electron capture cross sections. If the pro-

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jectile velocity v is greater than the orbital velocity of the electron being captured an atomic representation is usually justified. Then the wave functions $\varphi_{nlm}(\vec{r})$ or $\tilde{\varphi}_{nlm}(\vec{q})$ appear explicitely in the transition amplitude. The dominant contributions arise from $\vec{q}$ values peaked around $\vec{v}$. Classically, the longitudinal and transverse momentum transfers at the impact parameter b are $q_{\parallel} \approx v$ and $q_{\perp} \approx 2Z/(vb)$ so that $\sin(\frac{1}{2}x) \lesssim 2q_n Z/bv^3$ rapidly becomes small with increasing projectile velocity v. It is then a good approximation to put $d_n(x) = 1$ everywhere since only the region around $x = 0$ contributes to the transition. The resulting factorization of the density matrix (4a) leads to a simple prescription which drastically simplifies the computation of electron capture into arbitrary principal shells: *The total cross section for electron capture into the nth principal shell of the projectile with charge Z can be calculated as the capture into the 1s state of a substitute projectile with charge Z/n via the replacement*

$$\varphi_n^{\mathrm{eff}}(z) = n\varphi_{1s}(Z/n). \tag{5}$$

The replacement of wave functions is equally valid in momentum and in coordinate space.

As an illustration and application, we calculate the total cross section for Reaction (1) in the eikonal approximation.[11,12] We notice that for capture into high-Z projectiles high n will dominate and hence the initial and final bound states will be almost orthogonal. The problem of whether or not to include the internuclear interaction in the transition amplitude will then not arise. Let $\vec{r}$, $\vec{r}_{\mathrm{H}} = \vec{r} + \alpha\vec{R}$, and $\vec{r}_A = \vec{r} + (1-\alpha)\vec{R}$ denote the position of the electron with respect to the center of mass, the proton, and the projectile nucleus, respectively, with $\alpha = M_A/(M_A + M_{\mathrm{H}})$. The projectile follows a straight-line trajectory $\vec{R} = \vec{b} + \vec{z}_R$ with respect to the target nucleus. A sum rule similar to Eq. (4) can be applied if we use the eikonal approximation in its *prior* form for the transition amplitude

$$A(\mathrm{b},v) = -i\int_{-\infty}^{\infty} dt \langle \Psi_f^- | (-z/r_A) | \Psi_{1s} \rangle. \tag{6}$$

The time-dependent wave functions including translation factors[1] are given by

$$\Psi_{1s} = \varphi_{1s}(\vec{r}_{\mathrm{H}}) \exp(-i\epsilon_{\mathrm{H}} t) \exp(-i\alpha\vec{v}\cdot\vec{r} - \tfrac{1}{2}i\alpha^2 v^2 t) \tag{7a}$$

and

$$\Psi_f^- = \varphi_{nlm}(\vec{r}_A)\exp(-i\epsilon_A t) \exp[i(1-\alpha)\vec{v}\cdot\vec{r} - \tfrac{1}{2}i(1-\alpha)^2 v^2 t] \exp(-i\int_t^{\infty} dt/r_{\mathrm{H}}). \tag{7b}$$

The last factor represents the eikonal phase in its *prior* form.[13] Inserting Eqs.(7) into Eq. (6) we observe that the function $\varphi_{nlm}(\vec{r}_A)/r_A$ is the only term explicitely containing the coordinate $\vec{r}_A$. This suggests, in analogy to Eq. (2), to introduce the Fourier transform $g_{nlm}(\vec{q})$ by

$$r_A^{-1}\varphi_{nlm}(\vec{r}_A) = (2\pi)^{-3}\int d^3q\, g_{nlm}(\vec{q})\exp(-i\vec{q}\cdot\vec{r}_A). \tag{8}$$

Eliminitating the Coulomb interaction $1/r_A$ with the aid of the Schrödinger equation[6] one obtains a relation analogous to Eq. (4)

$$\sum_{lm} g_{nlm}^*(\vec{q})g_{nlm}(\vec{q}') = \frac{16\pi q_n^3}{(q^2 + q_n^2)(q'^2 + q_n^2)} \frac{\sin nx}{n \sin x}. \tag{9}$$

In the further reduction[14] the eikonal phase is replaced by an integral representation given by Gau and Macek,[15] and for the resulting exponentials the closed-form Fourier representation is introduced simultaneously with Eq. (8). The integrations over $\vec{r}$ and $t = Z_R/v$ can then be carried out. It becomes clear that the argument $\vec{q}$ of $g_{nlm}(\vec{q})$ is almost completely fixed: $q_{\parallel} = \frac{1}{2}v + (1 - q_n^2)/2v$ and $q_{\perp} \approx 0$ because otherwise the rapid oscillations of the integrand give negligible contributions to $A(\vec{b},v)$. As a consequence, if $|A(\vec{b},v)|^2$ is formed and summed over all substates for a given n we have $\vec{q} \approx \vec{q}$ and may apply Eq. (9) with $d_n(x) = \sin nx/(n \sin x) = 1$. This is in line with the general arguments preceding Eq. (5).

It is possible to carry our all integrations without further approximations. The details will be published elsewhere.[14] As a result we obtain the total capture cross section

$$\sigma_{1s-n}(z,v) = \frac{\pi\eta}{\sinh(\pi\eta)} \exp[-2\eta\tan^{-1}(\tfrac{1}{2}v - \eta\epsilon_{AH})]\{\tfrac{23}{48} + (\tfrac{1}{6} + \tfrac{5}{6}\epsilon_{AH})\eta^2 + \tfrac{5}{12}\epsilon_{AH}^2\eta^4\}\sigma_{1s-n}^{\mathrm{OBK}}(Z,v). \tag{10}$$

Here, $\eta = 1/v$, $\epsilon_{AH} = \epsilon_A - \epsilon_{\mathrm{H}} = \frac{1}{2}(1 - Z^2/n^2)$, and $\sigma_{1s-n}^{\mathrm{OBK}}(Z,v)$ is the OBK cross section.[1,2] In the particular case $n = Z = 1$ we recover the result of Dewangan[11] who used the post form of the eikonal approximation. As is well known, for asymptotically high energies the second Born term becomes dominant[1,2] and hence Eq. (10) is no longer valid.

Equation (10) has a remarkable simplicity, comparable to that of the OBK formula. For practical applications, one has to sum Eq. (10) over a great number of principal shells which all contribute to the capture cross sections. It is useful to express the result in terms of a theoretical scaling factor α defined by

$$\begin{aligned}\sigma_{\text{capt}} &= \textstyle\sum_n \sigma_{1s\text{-}n}(Z,v) \\ &= \alpha(Z,v) \textstyle\sum_n \sigma_{1s\text{-}n}{}^{\text{OBK}}(Z,v).\end{aligned} \tag{11}$$

The numerical calculations show that the factor $\alpha(Z,v)$ is almost independent of the projectile charge Z (within ± 5% up to $Z=26$). The energy dependence of the scaling factor α (calculated for He^{++}) is shown in Fig. 1. Also shown are experimental points[16-21] obtained by dividing σ_{capt} (expt.) by $\sigma_{1s\text{-}n}{}^{\text{OBK}}$. For partially stripped projectiles effective charges have been used according to Olson and Salop[5] for calculating σ^{OBK}. The agreement with the theoretical prediction is quite satisfactory in view of experimental errors of typically 30%. It should be noticed that a sensitive linear plot has been used rather than the conventional logarithmic plot ranging over many orders of magnitude. Considering the perturbative approach underlying Eq. (10) it is surprising that satisfactory results are obtained for projectile charges as high as 25. More experimental data are needed to test the limitations of the present approach.

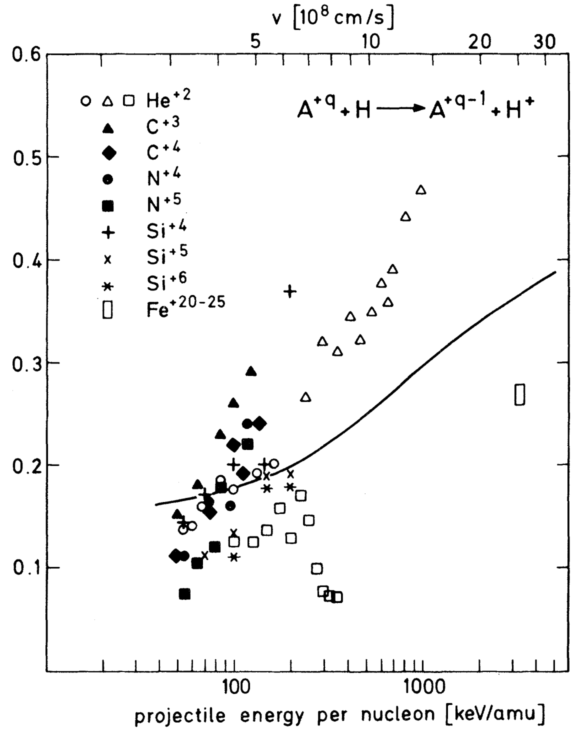

FIG. 1. The scaling factor α obtained by dividing capture cross sections by the corresponding OBK cross section is plotted as a function of projectile energy. The curve shows the theoretical result of Eq. (11). The points indicate experimental data with error bars of typically 30% omitted. Open circles, squares, and triangles are from Refs. 16, 17, and 18, respectively. The data for partially stripped ions C and N are from Ref. 19, for Si from Ref. 20. In all these cases, the effective charges g_{eff} given by Olson and Salop (see Refs. 5 and 20) have been used to calculate the OBK cross sections [$q_{\text{eff}}(\text{C}^{+3})=2.4$, $q_{\text{eff}}(\text{C}^{+4})=q_{\text{eff}}(\text{N}^{+4})=3.2$, $q_{\text{eff}}(\text{N}^{+5})=4.1$, $q_{\text{eff}}(\text{Si}^{+4})=3.0$, $q_{\text{eff}}(\text{Si}^{+5})=4.1$, $q_{\text{eff}}(\text{Si}^{+6})=5.1$]. The data for Fe^{+q} with $q=20\text{–}25$ are from Ref. 21. All six experimental points fall into the bar indicated in the figure if $Z=q$ is used to calculate σ^{OBK}. The agreement is actually better than shown here if comparison is made with $\alpha(Z\approx 25)$. The data of Refs. 17, 18, and 21 refer to H_2 and have been divided by a factor of 2 for a reduction to atomic hydrogen.

So far, we consider Eq. (10) as an explanation of the empirical scaling rule. We therefore propose that it should replace the corresponding OBK formula as a first and simple starting point for the estimation of total capture cross sections. We also wish to emphasize that Eq. (10) is just an illustration—as is the OBK formula[1,2]—for the use of the replacement rule (5). Other applications of this rule or the underlying representation (4) of the density matrix are certainly conceivable.

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Melting and Staging in Graphite Intercalated with Cesium

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The order-disorder transition in Cs-intercalated graphite has been studied using x-ray scattering. In C_8Cs the Cs layer transforms into a liquidlike phase in which the average Cs-Cs separation is incommensurate with the carbon net. Fundamentally different behavior is observed in a Cs-deficient stage-1 sample where commensurate "lattice-gas" melting occurs. Evidence is presented which shows that in low-stage material, staging is closely related to the melting transition in support of the model of Daumas and Hérold.

It is well known that certain chemical species form pure-stage graphite intercalation compounds which exhibit regular layer-stacking sequences along the *c* axis. In an elegant series of x-ray diffraction studies,[1] Parry and co-workers showed that whereas stage-1 C_8M (M = K, Rb, Cs) is three-dimensionally ordered at room temperature, nth-stage compounds $C_{12n}M$ ($n \geq 2$) contain two-dimensionally disordered M layers which order (or freeze) at T = 98, 159, and 163 K for M = K, Rb, and Cs, respectively. More recently, Onn, Foley, and Fischer[2] have studied the temperature-dependent resistivity of $C_{12n}M$ (n = 2, 3) and have observed anomalies which indicate the presence of two transitions. While stage-2 and higher-stage alkali intercalates have been extensively studied, there has been to date no quantitative analysis of the order-disorder phase transition in graphite intercalates. In addition, with the exception of a preliminary report by Ellenson *et al.*,[3] melting of the M layers in stage-1 compounds has not been addressed. Moreover, the relationship between melting and staging in graphite intercalates has not been examined.

In this Letter we report x-ray-diffraction studies of stage-1 C_8Cs and of unusual stage-1–stage-2 mixtures. We will show that Cs layers in C_8Cs melt three-dimensionally and that the melting is dependent on the vapor pressure of the Cs surrounding the sample. The melting transition is either quasi second order and of the lattice-gas type, in which Cs atoms commensurately occupy preferred carbon hexagon sites, or is first order and incommensurate. To verify these points we will present data for the liquid structure factors of the melted layers. We will also discuss the applicability of the four-state Potts model[4] to the second-order lattice-gas transition. Finally, we will show that melting and staging are closely related phenomena in graphite intercalates and we will present direct evidence in support of a model for staging which, though proposed several years ago by Daumas and Hérold,[5] has received little attention.

Samples were prepared from highly oriented pyrolytic graphite (HOPG) using the two-bulb method of Hérold.[6] $8 \times 8 \times 0.5\text{-mm}^3$ pieces of freshly cleaved HOPG were sealed in evacuated

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