The different effect of electron-electron interaction on the spectrum of atoms and quantum dots*

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Abstract. Though atoms and quantum dots typically contain a comparable number of electrons, the number of discrete levels resolved in spectroscopy experiments is very different for the two systems. In atoms, hundreds of levels are observed while in quantum dots that number is usually smaller than 10. In the present work, this difference is traced to the different confining potentials in these systems. In atoms, the soft confining potential leads to large spatial extent of the excited electron's wave function and hence to weak Coulomb interaction with the rest of the atomic electrons. The resulting level broadening is smaller than the single particle level spacing and *decreases* as the excitation energy is increased. In quantum dots, on the other hand, the sharp confining potential results in electron-electron scattering rates that grow rapidly with energy and fairly quickly exceed the approximately constant single particle level spacing. The number of discrete levels in quantum dots is hence limited by electron-electron interaction, whose effect is negligible in atoms.

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1 Introduction

This work compares the effect of electron-electron (e-e) interaction on the spectrum of atoms and quantum dots (QDs). It is motivated by the observation that although a large atom and a QD may contain a comparable number of electrons, their spectral characteristics are very different. In QDs, level broadening due to e-e interaction severely limits the number of observable discrete single particle excitations. In atoms on the other hand, hundreds of spectral lines have been measured and tabulated. Level widths are mainly due to electron-photon interaction, while the effect of e-e interaction is negligible. This work elucidates what differences between atoms and QDs account for their distinct spectral characteristics with respect to e-e lifetime.

Atoms and QDs both contain a comparable number of interacting electrons in a confined volume, while differing in size, symmetry and confining potential. QDs are typically larger, possess no particular symmetry, and are confined by a sharper potential. We find the difference in the spectral characteristics of these two systems is primarily due to the effect of the different confining potentials on e-e interaction.

We focus on single particle excitations in which one electron is excited to a weakly bound state, while the other electrons remain in their ground state configuration. We define a perturbation Hamiltonian corresponding to the residual interaction between the excited electron and the other electrons, and use second order perturbation theory to estimate e-e lifetimes. The effect of e-e interaction on the atomic spectrum is found to depend on the number of valence electrons. Alkali atoms, with a single valence electron, suffer no broadening. In complex atoms, single particle excitations may acquire a finite width. However, this width is smaller than the single particle level spacing and therefore does not limit the number of discrete single particle excitations. Interestingly, we find that e-e scattering rates in complex atoms decrease with increasing excitation energy.

QDs exhibit different behavior. Level broadening due to e-e interaction limits the number of observable single particle excitations. For example, Sivan *et al.* [1] were able to resolve only ~ 10 discrete levels in the excitation spectrum of a diffusive QD containing about 4000 electrons. Theoretical results [2] indicate that the number of observable discrete levels in the spectrum of a QD, *i.e.* the number of levels for which the e-e broadening is smaller than the single particle level spacing, is relatively small. In addition, e-e scattering rates in QDs are found to increase with excitation energy in a Fermi-liquid-like behavior.

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* The subject of the present paper was posed by the late Arkadi Aronov in the form of the following question "What is the difference between an uranium atom and a quantum dot?"

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The different effect of e-e interaction in complex atoms and QDs can be qualitatively understood in the following way. The scattering rate is proportional to the residual interaction matrix element squared times the density of final states. In QDs, due to the sharp confining potential, the matrix elements are on the average only weakly dependent on energy. The density of final states is proportional to the excitation energy squared. The resulting scattering rates grow with energy and eventually exceed the fairly constant single particle level spacing. In complex atoms, due to the shallow confining potential, the radius of the excited electron's orbit grows rapidly with energy leading to suppressed matrix elements and diminishing e-e scattering rates. The decrease of e-e scattering rates with increasing excitation energy is in sharp contrast to the corresponding trend in QDs and Fermi liquids. The hydrogen-like single particle level spacing also decreases with energy, but slower than the scattering rates. Consequently, in complex atoms, unlike QDs, the discrete nature of the spectrum is preserved.

The outline of this paper is as follows. In Section 2 we define e-e lifetime of a single particle excitation and discuss its meaning in a finite system. We consider in particular the manifestation of e-e lifetime in a spectroscopic measurement. In Section 3 we estimate the e-e lifetime of single particle excitation in atoms using well established atomic theory. We distinguish between alkali atoms and complex atoms. In Section 4 we review known results for e-e lifetime in QDs. Finally, in Section 5 we compare the results concerning e-e lifetime in atoms and in QDs.

2 Electron-electron lifetime and its manifestation in the measured spectrum

Consider an isolated, N-electron system in its ground state, $|\text{g.s.}\rangle$, and an excited state,

$$|\Psi(t=0)\rangle = c_i^\dagger c_j |\text{g.s.}\rangle, \tag{1}$$

where $c_i^\dagger$ and c_j are single particle creation and annihilation operators respectively. The e-e lifetime, denoted by τ_{ee}, is defined as the decay time of the initial state,

$$|\langle \Psi(t)|\Psi(t=0)\rangle|^2 \sim e^{-\frac{t}{\tau_{\text{ee}}}}. \tag{2}$$

This analysis assumes zero temperature. Generalization to finite temperature is straightforward.

The meaning of e-e lifetime in a system characterized by a discrete energy spectrum, such as an atom below the ionization threshold or a QD, should be clarified. Let $\{|\psi_\alpha\rangle\}$ be a basis of exact eigenstates of the system with energies $\{E_\alpha\}$. The initial state can be expressed as a superposition of these eigenstates,

$$|\Psi(t=0)\rangle = c_i^\dagger c_j |\text{g.s.}\rangle = \sum_\alpha \lambda_\alpha |\psi_\alpha\rangle \tag{3}$$

$$\sum_\alpha |\lambda_\alpha|^2 = 1.$$

The probability to remain in the initial state at time t is,

$$|\langle \Psi(t=0)|\Psi(t)\rangle|^2 = \left| \sum_\alpha |\lambda_\alpha|^2 e^{iE_\alpha t/\hbar} \right|^2. \tag{4}$$

Strictly speaking, as opposed to the case of a continuous spectrum, here there is no decay of the probability to remain in the initial state. This probability oscillates, and for long enough times it can get arbitrarily close to 1 (it can return exactly to 1 if the energy spectrum is commensurate). Nevertheless, as the number of dominant terms in (3) increases, the time it takes for the initial state to reconstruct becomes extremely long. On shorter time scales, which are our focus here, the probability effectively decays exponentially with time. Consequently, we define the e-e lifetime in a finite system as in (2), keeping in mind the restriction on the time scales ($t, \tau_{\text{ee}} \ll$ reconstruction time of the initial state).

The full interacting Hamiltonian H is divided into an unperturbed Hamiltonian H_0, which may include part of the e-e interaction (for example, the Hartree or Hartree-Fock part), and a perturbation Hamiltonian H_{int} which includes the rest of the e-e interaction. Following [4], we arrange the eigenstates of the unperturbed Hamiltonian H_0 in the form of a hierarchal tree emanating from the ground state of H_0. The first generation includes all eigenstates of H_0 which are coupled by H_{int} to the ground state, in particular single particle excitations (as in (1)) are included. Each generation includes all eigenstates of H_0 not already incorporated in the tree which are connected by non-vanishing interaction matrix elements to the previous generation. The problem of e-e lifetime of single particle excitations can be mapped on the problem of Anderson localization on the hierarchal tree, described by a Hamiltonian H_A. Each eigenstate of H_0 is a site in the hierarchal tree with an on-site energy equal to its eigenenergy. The hopping amplitude between two sites is given by the corresponding residual interaction matrix element. The single particle spectrum of H_A on the hierarchal tree is equivalent to the many-body spectrum of H. An excitation of the type (1) corresponds to a single site on the lattice. The time evolution of this site is determined by its overlap with exact eigenstates of H_A. An overlap of this site with extended states leads to exponential decay, while overlap limited to localized states leads to beating of few frequencies.

We now turn to discuss how e-e lifetime of single particle excitations is manifested in the measured many-electron spectrum of a quantum dot or an atom. A finite, isolated, Fermi system has discrete energy levels of zero width, which correspond to *exact* many-body eigenstates of the system. A spectroscopic measurement involves coupling the system to some external measuring device. Typically, the measurement operators are single electron ones. As a result, only many-body eigenstates with finite overlap with single particle excitations can be detected.

Consider a specific example, an optical absorption experiment on a many-electron system in its ground state

|g.s.⟩. The measurement operator in this case is given by,

$$O = \sum_{i,j} \lambda_{ij} c_i^\dagger c_j + \text{h.c.} , \qquad (5)$$

where λ_{ij} are coefficients (including matrix elements of photon operators) determining the strength of the coupling to various single particle excitations. The absorption of the system is characterized by the spectral function,

$$\alpha(\omega) = \sum_f |\langle f| \sum_{i,j} \lambda_{ij} c_i^\dagger c_j + \text{h.c.}|\text{g.s.}\rangle|^2 \delta(E_{\text{gs}} + \hbar\omega - E_f), \qquad (6)$$

where $|f\rangle$ and E_f are exact final many-body eigenstates and eigenenergies of the system, and E_{gs} is the ground state energy of the system. In the absence of e-e interaction, $\alpha(\omega)$ exhibits a series of δ-peaks corresponding to single particle excitations of the system. When e-e interaction is included, each single particle excitation becomes a superposition of several exact many-body eigenstates. The spectral function then displays many more absorption peaks corresponding to many-body eigenstates that overlap with single particle excitations. A many-body eigenstate can overlap several single particle excitations, which then generate interfering contributions to its absorption peak.

A significant simplification of the spectral function and its relation to the concept of e-e scattering rate is obtained by assuming that each many-body state $|f\rangle$ overlaps with at most one single particle excitation, an approximation equivalent to the reduction of the hierarchal tree into a Cayley tree [4]. This approximation is valid when the *intra*-generation matrix elements are negligible, or when the single particle level spacing is much larger than the resulting e-e broadening. Within this approximation, the intensity of peaks having significant overlap with a particular single-particle excitation $c_{i_0}^\dagger c_{j_0}|\text{g.s.}\rangle$ is approximated by $|\lambda_{i_0 j_0}|^2 |\langle f|c_{i_0}^\dagger c_{j_0}|\text{g.s.}\rangle|^2$. The relative intensity of all δ-peaks $|f\rangle$ associated with this single particle excitation is proportional to $|\langle f|c_{i_0}^\dagger c_{j_0}|\text{g.s.}\rangle|^2$.

Consider the time evolution of a system initially in a state, $|\psi(t=0)\rangle = c_{i_0}^\dagger c_{j_0}|\text{g.s.}\rangle$,

$$|\psi(t)\rangle = \sum_f e^{iE_f t/\hbar} \langle f|c_{i_0}^\dagger c_{j_0}|\text{g.s.}\rangle|f\rangle. \qquad (7)$$

The e-e scattering rate is determined by the energy spread of exact many-body eigenstates, $|f\rangle$, that participate in the sum (7), provided there are many such states. These many-body eigenstates are exactly those that generate the absorption peaks associated with $c_{i_0}^\dagger c_{j_0}|\text{g.s.}\rangle$ in the spectral function. Thus, e-e scattering rate of a single particle excitation is manifested in the energy width of the cluster of absorption peaks associated with it in the spectral function.

In a real experiment the resolution of the measuring device is finite. The measured many-body spectrum is smeared, so that each δ-peak in the spectral function appears as a broadened peak. This broadening may have a typical scale larger than the many-body level spacing. In this case, the many-body eigenstates can no longer be resolved in the spectrum. Then, a cluster of δ-peaks associated with a given single particle excitation appears as a broad single particle resonance whose width is equal to the inverse e-e lifetime of the excitation. Single particle resonances can be resolved as long as their broadening is smaller than the single particle level spacing. When the broadening of single-particle resonances exceeds the single-particle level spacing, the measured spectrum becomes essentially continuous.

3 E-e lifetime in atoms

Consider an atom composed of a fixed nucleus with charge Z and N electrons. The full non-relativistic Hamiltonian is given by,

$$H = \sum_i \left(\frac{p_i^2}{2m} - \frac{Ze^2}{r_i}\right) + \sum_{i \neq j} \frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} , \qquad (8)$$

where $\mathbf{p}_i$ and $\mathbf{r}_i$ are the momentum and position operators of the electrons, respectively, m is the electron's mass and e is the electron's charge.

E-e interaction has a significant effect on the atomic eigenstates and eigenenergies as demonstrated in the numerical calculations of a Cerium atom by Flambaum *et al.* [3]. Consequently, e-e lifetime can not be calculated using a non-interacting model and low order perturbation theory in the e-e interaction. One may use low order perturbation theory only if most of the e-e interaction is included in the unperturbed Hamiltonian.

We consider the e-e lifetime of highly excited single particle excitations (but still below the ionization threshold[1]), for which the excited electron spends most of the time in regions where the density of the other electrons is exponentially small. The excited electron may then be considered distinguishable from the other electrons. Thus, the atom is divided into a distinguishable excited electron and an ion containing all other electrons and the nucleus. The Hilbert space is spanned by direct products of the ionic states and the excited electron states.

The unperturbed Hamiltonian is given by,

$$H_0 = H_{\text{ion}} + h_{\text{e}} ; \quad h_{\text{e}} = \frac{p^2}{2m} + V_{\text{e}}(r).$$

H_{ion} is the full Hamiltonian of the ion, and h_{e} is the effective single particle Hamiltonian of the excited electron which includes an effective potential V_{e} induced by the nucleus and the spherically averaged ionic ground state electron density. In regions exterior to the ionic electron

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[1] Excitations above the ionization threshold are coupled by the free electromagnetic field to a continuous spectrum of an ion and an unbound electron, leading to auto-ionization. Auto-ionization processes dominate the e-e lifetime above the threshold.

density, the effective potential experienced by the excited electron is simply $V_e(r) \simeq -e^2/r$.

The perturbation Hamiltonian is,

$$H_{\mathrm{int}} = e^2 \int \mathrm{d}\mathbf{r}' \frac{\delta\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \qquad (9)$$

$$\delta\rho(\mathbf{r}') = \rho(\mathbf{r}') - \frac{1}{4\pi} \int \mathrm{d}\Omega \langle \mathrm{g.s}|\rho(\mathbf{r}')|\mathrm{g.s.}\rangle, \qquad (10)$$

where $\rho(\mathbf{r}')$ is the density operator of the $N-1$ electrons of the ion, and $\delta\rho(\mathbf{r}')$ is its fluctuating part. $|\mathrm{g.s}\rangle$ refers to the ionic ground state, and $\mathbf{r}$ is the position operator of the excited electron.

The golden rule is employed to calculate the e-e lifetime of such single particle excitations due to the residual coupling, H_{int}. We first consider the relevant density of states and the interaction matrix elements. The results are then used to estimate the e-e lifetime.

3.1 Density of states of H_0

The density of states of H_0, $G(E)$, is a convolution of the density of states of the excited electron, $g_e(E)$, and the density of states of the ion, $g_{\mathrm{ion}}(E)$,

$$G(E) = \int \mathrm{d}\epsilon\, g_e(\epsilon) g_{\mathrm{ion}}(E - \epsilon), \qquad (11)$$

where all energies are measured relative to the ground state.

The characteristics of $G(E)$ vary considerably between atoms. This is mainly due to $g_{\mathrm{ion}}(E)$ which strongly depends on the number of valence electrons and the number of available single particle states. The density of states of the excited electron, $g_e(E)$, depends only weakly on the specific atom under consideration; apart from the few lowest levels, the spectrum is hydrogen-like in all atoms. We discuss two limiting cases – alkali atoms characterized by a sparse spectrum in the vicinity of the ground state, and complex atoms characterized by an exponentially large density of states in that energy range.

Alkali atoms have a single valence electron in an s-shell. All other electrons reside in closed shells with relatively low single particle energies. Exciting an electron from these inner shells has a large energy cost. Hence, the ionic spectra are characterized by a large gap (nearly $\sim E_i$) at the ground state.

Atoms with a few valence electrons are referred to as complex atoms. Again, core electrons are ineffective in e-e scattering, and only valence electrons contribute to the level width of the excited electron states. Typically complex atoms have several open shells with similar single particle energies. As Z increases, these shells can acquire large orbital momenta, and consequently large degeneracy. The distribution of $N_{\mathrm{val}} - 1$ ionic valence electrons among those N_s single particle states gives rise to $\sim N_s^{N_{\mathrm{val}}-1}$ many-body states of comparable energy, and to an average many-body level spacing, $\Delta \sim N_s^{-(N_{\mathrm{val}}-1)} e^2/a_0$. Thus,

the many-body density of states in complex atoms is exponentially large even in the vicinity of the ground state. Generally e-e interaction (a) lifts most degeneracies peculiar to the non-interacting spectrum, leading to a more homogeneous spectrum and (b) mixes Slater determinants, so that exact eigenstates are superpositions of many Slater determinants [3]. These effects enhance the density of final states since the single particle selection rules are relaxed.

3.2 Interaction matrix elements

We consider matrix elements of H_{int} between the initial excited state, $|i\rangle = |n_i, l_i, m_i\rangle|\alpha_i\rangle$, and any final state, $|f\rangle = |n_f, l_f, m_f\rangle|\alpha_f\rangle$. In our notation, $|n_i, l_i, m_i\rangle$ and $|n_f, l_f, m_f\rangle$ refer to the initial (hydrogen-like) and final states of the excited electron[2], respectively. Similarly, $|\alpha_i\rangle$ and $|\alpha_f\rangle$ refer to the initial and final states of the ion. $|\alpha_i\rangle$ and $|\alpha_f\rangle$ are exact eigenstates of the ion, so that $|i\rangle$ and $|f\rangle$ are eigenstates of H_0.

One can expand the interaction Hamiltonian, H_{int}, in multipoles. Since the excited electron is practically exterior to the atom this multipole expansion can be approximated by,

$$H_{\mathrm{int}} \simeq \sum_{k=0}^{\infty} \sum_{m_k=-k}^{k} \frac{1}{2k+1} \frac{1}{r^{k+1}} Y_{k,m_k}(\theta, \phi)$$

$$\times \int \mathrm{d}\mathbf{r}'\, r'^k \delta\rho(\mathbf{r}') Y_{k,m_k}^*(\theta', \phi'). \qquad (12)$$

The interaction Hamiltonian in this approximation is a sum of terms acting separately on the excited electron and on the ion. Equation (12) is essentially an expansion in the ratio between the characteristic radius of $\delta\rho(\mathbf{r})$ ($\sim a_0$) and the average radius of the excited electron ($\sim n^2 a_0$). This ratio is small due to the large spatial extent of the excited electron, which follows directly from the softness of the atomic confining potential at large distances. Roughly, we expect the kth term in the sum to scale with n as $\sim 1/n^{2(k+1)}$. Thus, high order terms in the expansion diminish quickly, and the dipole term ($k=1$) is dominant (unless it vanishes).

Calculation of the interaction matrix elements in this approximation involves separate calculations of matrix elements for the excited electron, $\langle n_i, l_i, m_i|\frac{1}{r^{k+1}}Y_{k,m_k}|n_f, l_f, m_f\rangle$, and for the ion, $\int \mathrm{d}\mathbf{r}'\, r'^k Y_{k,m_k}^*(\theta', \phi')\langle \alpha_i|\delta\rho(\mathbf{r}')|\alpha_f\rangle$. The infinite sums in (12) contain only few non-vanishing terms, due to angular momenta selection rules[3].

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[2] We do not specify explicitly the spin quantum number since it is unaffected by the interaction.

[3] The sum over m_k reduces to a single term with $m_k = m_f - m_i$. As a result of parity conservation, the sum over k has non-vanishing terms for either odd or even values of k (but not for both). The $k=0$ term (monopole) vanishes since it is already included in H_0. In addition k is upper bounded by $l_i + l_f$ due to angular momenta addition rules for the excited electron.

Consider the dominant dipole term ($k = 1$). The matrix element for the excited electron is given by,

$$\langle n_i, l_i, m_i | \frac{Y_{1,m}}{r^2} | n_f, l_f, m_f \rangle \qquad (13)$$

$$= \frac{2}{e^2 a_0} \frac{\epsilon_i - \epsilon_f}{l_i(l_i+1) - l_f(l_f+1)} \langle n_i, l_i, m_i | Y_{1,m} | n_f, l_f, m_f \rangle.$$

This term vanishes unless $l_f = l_i \pm 1$. For hydrogen-like wavefunctions,

$$\langle n_i, l_i, m_i | \frac{Y_{1,m}}{r^2} | n_f, l_i \pm 1, m - m_i \rangle \qquad (14)$$

$$\simeq \frac{1}{a_0^2} \frac{1}{n_i^3 l_i} \langle l_i, m_i | Y_{1,m} | l_i \pm 1, m - m_i \rangle$$

$$\times \int dr \, u_{n_i, l_i}(r) u_{n_f, l_i \pm 1}(r),$$

where $u_{n,l}$ are the radial wavefunctions. The overlap integral and hence the matrix element are strongly suppressed as n_f differs from n_i.

Calculation of the ionic part of the interaction matrix elements is notably more difficult, since it depends on the ionic many-body wavefunctions. Due to parity conservation, the diagonal matrix elements of the dipole term vanish, so that it does not couple degenerate states. The non-diagonal matrix elements are given by,

$$\int d\mathbf{r} \, r Y_{1,m}^*(\theta, \phi) \langle \alpha_i | \delta\rho(\mathbf{r}) | \alpha_f \rangle \qquad (15)$$

$$= \langle \alpha_i | \int d\mathbf{r} \, r Y_{1,m}^*(\theta, \phi) \rho(\mathbf{r}) | \alpha_f \rangle = \langle \alpha_i | d_m^1 | \alpha_f \rangle,$$

where $\{d_m^1\}_{m=\pm 1,0}$ is the tensorial representation of the dipole operator, $\mathbf{d} = \int d\mathbf{r} \, \mathbf{r} \rho(\mathbf{r})$.

The dipole operator can be expressed in terms of single particle creation and annihilation operators,

$$\mathbf{d} = \sum_{i,j} \mathbf{d}_{ij} c_i^\dagger c_j, \qquad (16)$$

where $\mathbf{d}_{ij}$ are the matrix elements of the single particle dipole operator. This sum is conveniently decomposed as follows,

$$\mathbf{d} = \sum_{i,j \in \text{open shells}} \mathbf{d}_{ij} c_i^\dagger c_j + \sum_{i \text{ or } j \in \text{core}} \mathbf{d}_{ij} c_i^\dagger c_j. \qquad (17)$$

We define a projected dipole operator, $\mathbf{d}_{\text{proj}} = \sum_{i,j \in \text{open shells}} \mathbf{d}_{ij} c_i^\dagger c_j$. Then,

$$\sum_f |\langle \alpha_f | \mathbf{d} | \alpha_i \rangle|^2 = \langle \alpha_i | \mathbf{d}_{\text{proj}}^2 | \alpha_i \rangle, \qquad (18)$$

where the sum is restricted to final ionic states with no core excitations. The last equation can be used to estimate the average squared dipole matrix element appearing in the golden rule,

$$\overline{|\langle \alpha_f | \mathbf{d} | \alpha_i \rangle|^2} \sim \frac{\langle \alpha_i | \mathbf{d}_{\text{proj}}^2 | \alpha_i \rangle}{N_{\text{tot}}}, \qquad (19)$$

where N_{tot} is the number of final ionic states coupled by the interaction to the initial state.

The diagonal matrix elements of $\mathbf{d}_{\text{proj}}$ are given by,

$$\langle \alpha_i | \mathbf{d}_{\text{proj}}^2 | \alpha_i \rangle = \int d\mathbf{r} \, r^2 \langle \alpha_i | \rho_{\text{proj}}(\mathbf{r}) | \alpha_i \rangle \qquad (20)$$

$$+ \int d\mathbf{r} \, d\mathbf{r}' \, \mathbf{r} \cdot \mathbf{r}' \langle \alpha_i | g(\mathbf{r}, \mathbf{r}') | \alpha_i \rangle.$$

where $\rho_{\text{proj}} = \sum_{i,j \in \text{open shells}} c_i^\dagger c_j$ and $g(\mathbf{r}, \mathbf{r}') \equiv \rho_{\text{proj}}(\mathbf{r}) \times \rho_{\text{proj}}(\mathbf{r}') - \delta(\mathbf{r} - \mathbf{r}') \rho_{\text{proj}}(\mathbf{r})$. The first term in (20) is due to auto-correlation while the second reflects correlations between different electrons. Since repulsive interaction tends to keep valence electrons away from each other, $g(\mathbf{r}, \mathbf{r}') < 0$ for small $|\mathbf{r} - \mathbf{r}'|$. Furthermore, for $|\mathbf{r} - \mathbf{r}'|$ larger than the average distance between electrons, $g(\mathbf{r}, \mathbf{r}')$ becomes very small. Consequently,

$$\langle \alpha_i | \mathbf{d}_{\text{proj}}^2 | \alpha_i \rangle \sim \langle \alpha_i | \int d\mathbf{r} \, r^2 \rho_{\text{proj}}(\mathbf{r}) | \alpha_i \rangle. \qquad (21)$$

This leads to the estimate,

$$\overline{|\langle \alpha_f | \mathbf{d} | \alpha_i \rangle|^2} \sim \frac{N_{\text{val}} R_{\text{val}}^2}{N_{\text{tot}}}, \qquad (22)$$

where R_{val} is the average radius of valence electrons. The number of valence electrons, as well as their average radius, vary from atom to atom.

Alternatively, $\langle \alpha_i | \mathbf{d}_{\text{proj}}^2 | \alpha_i \rangle$ can be estimated using the chaotic nature of the many-body eigenstates. $|\alpha_i\rangle$ are decomposed into a sum of single Slater determinants, $|\alpha_i\rangle = \sum_m \Delta_m |\delta_m\rangle$, where $\Delta_m \equiv \langle \delta_m | \alpha_i \rangle$. Following Flambaum [3] it is assumed that $|\alpha_i\rangle$ are chaotic superpositions, so the coefficients Δ_m are random. The number of significant components in the decomposition is denoted by $\mathcal{N}$, so that $|\Delta_m| \sim \frac{1}{\sqrt{\mathcal{N}}}$. Thus,

$$\langle \alpha_i | \mathbf{d}_{\text{proj}}^2 | \alpha_i \rangle = \sum_{m,n} \Delta_m^* \Delta_n \langle \delta_m | \mathbf{d}_{\text{proj}}^2 | \delta_n \rangle \qquad (23)$$

$$= \sum_m |\Delta_m|^2 \langle \delta_m | \mathbf{d}_{\text{proj}}^2 | \delta_m \rangle$$

$$+ \sum_{m \neq n} \Delta_m^* \Delta_n \langle \delta_m | \mathbf{d}_{\text{proj}}^2 | \delta_n \rangle.$$

It is easily shown that the matrix elements of $\mathbf{d}_{\text{proj}}^2$ in a basis of single Slater determinants are all $O(N_{\text{val}})$, where N_{val} is the number of valence electrons. The first sum in (23) is $O(N_{\text{val}})$, since it has $\mathcal{N}$ positive terms of order $\frac{1}{\mathcal{N}} O(N_{\text{val}})$. The second sum is of the same order, since it is a random sum of $\mathcal{N}^2$ terms of order $\frac{1}{\mathcal{N}} O(N_{\text{val}})$. This point of view leads again to (22).

A rough estimate of the interaction matrix element, taking into account the dominant dipole contribution (for $|i\rangle$ and $|f\rangle$ satisfying the dipole selection rules), is obtained

by combining (14) and (22),

$$|\langle i|H_{\text{int}}|f\rangle|^2 \sim \left(\frac{1}{3}\right)^2 \left(\frac{1}{a_0^2 n_i^3 l_i}\right)^2 \frac{e^4 N_{\text{val}} R_{\text{val}}^2}{N_{\text{tot}}} \quad (24)$$

$$\sim \frac{1}{N_{\text{tot}}} \left(\frac{e^2}{a_0}\right)^2 \frac{N_{\text{val}} R_{\text{val}}^2}{10(n_i^3 l_i)^2},$$

for n_f close to n_i. The dependence upon the specific atom under consideration comes from the ionic part of the matrix element *via* $N_{\text{tot}}, N_{\text{val}}$ and R_{val}. The dependence of the matrix elements on the initial excitation energy is mainly due to the excited electron. Since N_{tot} grows exponentially with N_{val}, the interaction matrix elements are very small compared with the level spacing of the single particle Hamiltonian, h_e.

3.3 E-e lifetime

3.3.1 Alkali atoms

Alkali atoms have a single valence electron ($N_{\text{val}} = 1$). The ionic spectrum is characterized by a large gap (on the order of the ionization energy) at the ground state, due to the large energy cost associated with exciting core electrons. The relevant density of states for scattering vanishes and no level broadening due to e-e interaction occurs. Exact low lying many-body eigenstates can be identified with particular single-particle excitations (*i.e.* there is a large overlap $\lesssim 1$ between these two states). The number of observable single particle excitations is very large, because the single particle spectrum becomes hydrogen-like as the ionization threshold is approached.

The suppression of e-e broadening is characteristic of systems with a gap at the Fermi level, and it persists to excitation energies considerably larger than the gap. The atomic system is unique in that the single particle spectrum becomes dense above the energy gap, so the number of single particle excitations in alkali atoms for which e-e broadening is suppressed is infinite in principle.

3.3.2 Complex atoms

We argued earlier that the density of states of ionic excitations in complex atoms is exponentially large. The question is whether this density of states is sufficient to induce broadening of single particle excitations. The general criteria for applying the golden rule is that the resulting scattering rate, $\hbar/\tau_{\text{ee}} \simeq \overline{|\langle H_{\text{int}}\rangle|}^2 G_{\text{final}}(E)$ (where $\overline{|\langle H_{\text{int}}\rangle|}$ is the average interaction matrix element and $G_{\text{final}}(E)$ is the relevant density of final states), is larger than the level spacing of the relevant final unperturbed states, $1/G_{\text{final}}(E)$. This requirement is equivalent to the condition,

$$\frac{1}{G_{\text{final}}(E)} < \overline{|\langle H_{\text{int}}\rangle|}. \quad (25)$$

$G_{\text{final}}(E)$ refers only to final states which are coupled to the initial state by the interaction, and generally may be considerably smaller than the full many-body density of states. In our case, $G_{\text{final}}(E)$ is smaller than the full many-body density of states due to selection rules associated with symmetry (for the dominant dipole term). Nevertheless, $G_{\text{final}}(E)$ still grows exponentially with the number of valence electrons N_{val}. N_{tot} is the number of accessible final states, and the relevant density of states is roughly given by $G_{\text{final}}(E) \simeq N_{\text{tot}} a_0/e^2$. We saw in the previous section that $\overline{|\langle H_{\text{int}}\rangle|} \propto 1/\sqrt{N_{\text{tot}}}$. Since $1/G_{\text{final}}(E) \propto 1/N_{\text{tot}}$, eventually for large enough N_{tot}, (25) is satisfied. We conclude that *for large enough N_{tot}, e-e interaction leads to finite lifetime of single particle excitations.*

The e-e lifetime of a single particle excitation $|i\rangle$ with energy E, according to Fermi's golden rule, is,

$$\frac{1}{\tau_{\text{ee}}} = \frac{2\pi}{\hbar} \sum_{n_f, l_f, m_f} g_{\text{ion}}(E + \frac{e^2}{2a_0 n_f^2})\overline{|\langle i|H_{\text{int}}|f\rangle|^2}. \quad (26)$$

Summation is restricted to final states of the excited electron, as we have averaged the interaction matrix elements over the ionic states and correspondingly introduced the ionic density of states. Taking into account dipole matrix elements only (Eq. (24)), one obtains,

$$\frac{1}{\tau_{\text{ee}}} \sim \frac{2\pi}{\hbar} \left(\frac{N_{\text{tot}} a_0}{e^2}\right) \left(\frac{e^4}{N_{\text{tot}} a_0^2} \frac{N_{\text{val}} R_{\text{val}}^2}{10(n_i^3 l_i)^2}\right) \quad (27)$$

$$\sim \frac{2\pi}{\hbar} \frac{e^2}{a_0} \frac{N_{\text{val}} R_{\text{val}}^2}{10(n_i^3 l_i)^2}.$$

We note that N_{tot} has cancelled out of the final expression for τ_{ee}[4]. The single particle excitation spectrum is hydrogen-like with $E_{n_i} \sim -e^2/2a_0 n_i^2$, so the single particle level spacing is $\propto 1/n_i^3$. For large enough n_i, the level spacing of the excited electron becomes smaller than the level spacing of the ion. For such single particle excitations, no e-e lifetime can be defined and (26) becomes irrelevant.

The calculated e-e scattering rate depends on the specific atom through $N_{\text{val}} R_{\text{val}}^2$, which depends irregularly on Z due to the shell structure. The number of valence electrons in atoms is < 14, and the resulting e-e scattering rates are much smaller than the single particle level spacings.

One may consider atoms with larger Z. According to the Thomas Fermi (TF) model (see Appendix A), $N_{\text{val}} \propto Z^{1/3}$ for small Z, but it saturates to a constant in the limit $Z \to \infty$. This yields e-e scattering rates which are independent of Z in this limit. The TF model, however, ignores the shell structure. The shell structure suggests that the largest open sub-shell in an atom may have

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[4] We note the analogy between our calculation and the calculation of the lifetime of an atomic level due to electron-photon interaction in a box of volume V. There, the interaction matrix elements are $\propto 1/\sqrt{V}$, while the density of states is $\propto V$. The volume in this calculation plays a role similar to N_{tot} in our calculation – for large enough volume, the golden rule is valid and the resulting lifetime is independent of V.

$Z^{1/3}$ electrons, so the number of valence electrons fluctuates, as a function of Z, between $O(1)$ and $O(Z^{1/3})$ electrons. The average radius of the valence electrons, $R_{\rm val}$, also depends on Z. As the number of valence electrons increases, we expect $R_{\rm val}$ to decrease because the screening of the nucleus is less efficient. This tends to reduce the fluctuations in $N_{\rm val}R_{\rm val}^2$ as a function of Z. Thus, e-e scattering rates in atoms fluctuate with Z. A small fraction of atoms have $O(Z^{1/3})$ valence electrons. For such atoms the typical scattering rates increase as a function of Z ($1/\tau_{\rm ee} \propto Z^\alpha$, $\alpha < 1/3$), and eventually, for large enough Z, become larger than the single particle level spacing. For most atoms, however, the number of valence electrons is small and the e-e scattering rates of single particle excitations remain smaller than the single particle level spacing.

For a specific complex atom, the width of single particle excitations decreases as the excitation energy increases. This decrease is a direct consequence of the soft atomic potential at large distances ($r \gg a_0$). A small increase in the excitation energy amounts to a substantial increase of the average radius of the excited electron. As a result, the interaction matrix elements and therefore the e-e broadening are significantly reduced.

4 E-e lifetime in quantum dots

In this section we review results of calculations of e-e lifetime in QDs. We distinguish between ballistic QDs, in which the mean free path, l, is larger or equal to the linear dimension of the QD, L, and diffusive QDs, in which $l \ll L$. In both cases the non-interacting ground state is a filled Fermi sphere. Single particle excitations are broadened by e-e interaction which induces scattering to 2-electron 1-hole excitations. In the ballistic regime, these final states are restricted by momentum and energy conservation. In the diffusive regime, the constraint of momentum conservation is relaxed. The available phase space for scattering increases, leading to larger e-e scattering rates.

Single particle resonances can be observed as long as their width, $\hbar/\tau_{\rm ee}$, is smaller than the single particle level spacing, δ. The e-e scattering rates are used to estimate the number of resolvable single particle resonances. The results show that e-e lifetime severely limits the number of discrete single particle excitations in the measured spectrum of QDs in both the ballistic and the diffusive regimes.

The e-e lifetime in ballistic QDs is calculated in the standard Fermi liquid model [2]. The resulting e-e lifetime is given by[5],

$$\frac{\hbar}{\tau_{\rm ee}} \simeq \frac{E^2}{\epsilon_f}, \tag{28}$$

where ϵ_f is the Fermi energy. The e-e scattering rates increase with excitation energy and become comparable to

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[5] There is a logarithmic correction to this result in 2-D.

the single particle level spacing for $E \sim \sqrt{\delta\,\epsilon_f}$. The number of resolvable single particle excitations is therefore,

$$\mathfrak{N} \simeq \sqrt{\frac{\epsilon_f}{\delta}} \simeq \sqrt{N} \tag{29}$$

where $N \gg 1$ is the number of electrons in the QD.

The e-e lifetime of a single particle excitation in a diffusive QD for $E < E_c$ is given by [2],

$$\frac{\hbar}{\tau_{\rm ee}} \simeq \frac{160\delta}{\pi} \left(\frac{E}{E_c}\right)^2 \tag{30}$$

where $E_c = 4\pi^2 \hbar D/L^2$ is the Thouless energy and D is the diffusion coefficient. The number of resolvable single particle excitations in this case is,

$$\mathfrak{N} \sim \frac{E_c}{\delta} = g \tag{31}$$

where g is the dimensionless conductance. This number is typically very small (much smaller than $\sqrt{N}$ levels obtained in the ballistic case). This is consistent with the experimental results of Sivan *et al.* [1].

5 Discussion

The e-e lifetime of single particle excitations in atoms below the ionization threshold was estimated, and found to be small relative to the single particle level spacing. E-e interaction, hence, does not limit the number of observed discrete single particle excitations. In practice, broadening due to electron-photon interaction limits the number of resolved single particle excitations. In QDs, experimental and theoretical results indicate that e-e interaction limits the number of discrete single particle excitations in both the ballistic and the diffusive regimes. We also find that e-e scattering rates in atoms decrease with excitation energy (below the ionization threshold), in contrast to QDs.

The profoundly different e-e scattering rates in atoms and in QDs result from the different confining potentials. In atoms, the potential is very soft at large distances, $\sim 1/r$, while in QDs, it is characterized by sharper boundaries, $\sim r^2$. The confining potential determines the spatial extent of the excited single particle wavefunctions as well as the single particle spectrum. These properties have substantial influence on e-e scattering rates.

In QDs the confining potential generates a fairly uniform single particle spectrum in the vicinity of the Fermi level. The single particle spectrum leads to a relevant density of states (in this case, the 2-electron 1-hole density of states) which grows quadratically with excitation energy. The resulting e-e scattering rates increase with excitation energy. The spectrum consists of a small number of discrete single particle resonances, beyond which the broadening exceeds the single particle level spacing, and the single particle resonances merge and form a continuous spectrum.

In contrast, the Coulomb confining potential in atoms produces a dense single particle excitation spectrum

as the ionization threshold is approached. The spatial extent of an excited electron's wavefunction increases rapidly with excitation energy. The interaction matrix elements hence decrease with excitation energy leading to reduced e-e scattering rates, which do limit the number of observed discrete levels.

These results can be described using the Cayley tree picture discussed earlier (2). In both atoms and QDs one uses single electron measurement operators to probe the many-body spectrum of the system. However the nature of the excited states generated is different in these systems. Single particle excitations in atoms (below the ionization threshold) are localized on the Cayley tree, while in QDs nearly all single particle excitations correspond to extended states. The number of resolvable single particle excitations in atoms is therefore much greater than in QDs.

Appendix A: Calculation of the number of active electrons within the Thomas Fermi model

Electrons that participate in scattering processes contributing to the e-e lifetime of the excited electron are referred to as *active* electrons. Since core electrons have low single particle energies they can not participate in such scattering, and the number of active electrons is equal to the number of valence electrons. Within the TF model, the number of active electrons, N_{val}, is estimated from the single particle density of states.

The TF effective potential in a neutral atom, V_{TF}, is the solution of the TF equation,

$$\frac{\mathrm{d}^2 V(r)}{\mathrm{d}r^2} = -\frac{8\sqrt{2}}{3\pi}\frac{1}{e a_0^{3/2}}(\mu - V(r))^{3/2}, \qquad (32)$$

with $V(r \to 0) \sim -\frac{Ze^2}{r}$ and $V(r \to \infty) \to 0$. Here, μ is the chemical potential and $a_0 = \hbar^2/me^2$ is the Bohr radius. Using the TF potential, we can obtain the atomic single particle density of states in the semi-classical approximation, g_{TF}. Introducing explicit Z-dependence we have,

$$g_{\mathrm{TF}}(\epsilon, Z) = 2\frac{(2m)^{3/2}}{\pi\hbar^3}\int_0^{R(\epsilon,Z)} r^2\sqrt{(\epsilon - V_{\mathrm{TF}}(r,Z))}\,\mathrm{d}r \qquad (33)$$

where $R(\epsilon, Z)$ is the classically accessible radius given by $V_{\mathrm{TF}}(R(\epsilon, Z), Z) = \epsilon$.

The ionization energy of the atom, which is the difference between the chemical potential of a neutral atom ($\mu = 0$) and that of an ion ($N = Z - 1$), can be calculated within the TF approximation. To lowest order in $1/Z$, the ionization energy is constant for all atoms and given by [5],

$$E_{\mathrm{ion}}^{\mathrm{TF}} \approx 0.109\frac{e^2}{a_0}\,. \qquad (34)$$

This result does not account for the irregular variation of E_{ion} as a function of Z arising from the shell structure.

According to our definition, the single particle energy of active electrons has to be such that they can be excited above the atomic Fermi level by an energy of order E_{ion}. Thus, the number of active electrons in the TF atom is estimated by,

$$N_{\mathrm{val}}^{\mathrm{TF}}(Z) = \int_{-e^2/a_0}^0 g_{\mathrm{TF}}(\epsilon, Z)\mathrm{d}\epsilon. \qquad (35)$$

We find that only a small number of electrons ($\ll Z$) are active. The contribution of core electrons to $N_{\mathrm{val}}^{\mathrm{TF}}(Z)$ is $\sim Z^{-1/3}$, so for large atoms $N_{\mathrm{val}}^{\mathrm{TF}}$ is composed solely of electrons from the outer shell ($r \gg Z^{-1/3}a_0$).

The results of numerical calculation of $N_{\mathrm{val}}^{\mathrm{TF}}(Z)$ show that indeed $N_{\mathrm{val}}^{\mathrm{TF}}(Z) \ll Z$. For naturally occurring atoms ($Z \sim 100$), $N_{\mathrm{val}}^{\mathrm{TF}}$ is found to be roughly proportional to $Z^{1/3}$. However, within the TF model, $N_{\mathrm{val}}^{\mathrm{TF}}(Z)$ does not diverge as $Z \to \infty$. Rather it is upper bounded[6]. Hence, for large enough Z the number of active electrons in a TF atom is constant, independent of Z. There are, however, no real atoms with sufficiently large Z-values to check this prediction experimentally.

For a particular atom, the actual number of active electrons, $N_{\mathrm{val}}(Z)$, depends irregularly on Z due to the shell structure. It is $N_{\mathrm{val}}(Z)$ and not $N_{\mathrm{val}}^{\mathrm{TF}}(Z)$, which determines the characteristics of the spectra of different atoms. Nevertheless, $N_{\mathrm{val}}^{\mathrm{TF}}(Z)$ gives a rough estimate for $N_{\mathrm{val}}(Z)$, and leads us to the important observation that this number is $\ll Z$.

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[6] The exact value of the bound is meaningless since it depends on the definition of the relevant energy range. The important point is the existence of an upper bound that is independent of Z.