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LETTER TO THE EDITOR

Orthogonality catastrophe in Coulomb glass

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Abstract. It is shown that in the presence of weak quantum mechanical tunnelling, an orthogonality catastrophe type of effect is found in Coulomb glass systems in 3D and 2D, under the application of a long-ranged $1/r$ type potential. Some physical implications of the effect are discussed.

It is well known that the response of a non-interacting electron gas to the sudden application of an external potential is singular at long times (orthogonality catastrophe) (Anderson 1967, see also Mahan 1974). This phenomenon can be understood by considering the overlap integral between the two ground states before and after the external potential is switched on (Anderson 1967),

$$\langle G|G'\rangle = \det|A_{nn'}|_{E_n<E_f,E_{n'}<E_{f'}} \leqslant r \prod_{E_{n'}<E_{f'}} \left(1 - \sum_{E_n>E_f} |A_{nn'}|^2\right)^{1/2} \tag{1}$$

where E_f and $E_{f'}$ are the Fermi energies of the system in the *absence* and *presence* of the external potential, respectively, and n and n' are the corresponding one-particle eigenstates. $A_{nn'}$ is the overlap integral between the eigenstates n and n'. In metals, it is found that $\langle G|G'\rangle \to (1/N)^\alpha$ when $N \to \infty$, where N is the number of electrons present in the system and α is a positive number. This particular behaviour shows up in, for example, x-ray absorption experiments where the absorption spectrum exhibits a power law behaviour ω^α at the absorption threshold (see e.g. Mahan 1974, 1981). Physically, this behaviour occurs when the number of available states n,n' having appreciable magnitude of $|A_{nn'}|^2$ is large, so although individual values of $|A_{nn'}|^2$ may be small, the product of factors $(1 - \Sigma_{E_n>E_f}|A_{nn'}|^2)^{1/2}$ becomes vanishingly small when the number of electrons present in the system goes to infinity. For weak external perturbations the factor $|A_{nn'}|^2$ can be large only when the energies E_n and $E_{n'}$ are close in value, so that the dominating contributions are from low-energy particle–hole excitations under the application of the external potential.

The problem which interests us is whether this phenomenon is also found in Coulomb glass systems where electrons are strongly localized. For example, electrons in the impurity band of lightly-doped, compensated semiconductors or in amorphous semiconductors (see e.g. Pollak and Ortunö 1985 and Efros and Shklovskii 1985). In particular, we are interested in the situation in which the suddenly applied potential is of the form $v(r) = Ze^2/r$, i.e. a long-ranged Coulomb potential. This potential is interesting for two reasons:

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(i) because of the weak screening effect (see discussion below) it turns out that in a photo-emission experiment on deep core electronic states, the 'suddenly' appearing potential generated by the ionized atom is essentially an unscreened Coulomb potential, and

(ii) because electrons are localized the main effect of adding or removing an electron from a Coulomb glass system is the introduction of a Coulomb potential.

Therefore, this process also determines the one-particle properties of the Coulomb glass system.

This problem has been discussed by Efros and Shklovskii (1975, 1976, see also Baranovskii *et al* 1980) for the case in which electrons are completely localized (i.e. electron hopping is strictly prohibited). They found that the low energy behaviour of this 'classical' system is dominated by two features:

(i) the presence of a soft gap in the one-particle density of states (Coulomb gap), and

(ii) the existence of a *constant* density of states for particle–hole excitations (Efros and Shklovskii 1975, 1976, 1985, Davies *et al* 1984). We shall consider the more realistic situation in which a small quantum mechanical overlap integral $I(r) \sim I_0 e^{-r/a_0}$ also exists between different localized states separated by a distance r, where $a_0 \sim$ interatomic distance, so relaxation of the system by tunnelling is allowed in the presence of an external potential.

To describe the Coulomb glass system we follow the model introduced by Shklovskii and Efros (1981) in which electrons occupy sites of a periodic lattice of lattice constant a_0. There are twice as many sites as electrons, which are assumed to be spinless. Each site i is assigned a random energy φ_i, uniformly distributed in the interval $-A$ to A. Charge neutrality is maintained by assuming existence of a uniform, positive background of charge $+e/2$ per site. Electrons are allowed to hop between sites through a small nearest neighbour hopping term I_0. We shall consider the limit $e^2/a_0 \ll A$ and $I_0 \ll A$ so that electrons are strongly loalized with localization length $\sim a_0$.

The low energy excitations of this system are particle–hole pairs formed by moving electrons from occupied states to empty states (Efros and Shklovskii 1975, 1976, Shklovskii and Efros 1981). For excitations with energy $\leqslant \omega$, the typical distance between the sites of a pair is of order $r_\omega \sim a_0 \ln(2I_0/\omega)$ (Shklovskii and Efros 1981) and the average distance between two pairs is of order $a_0(A/\omega)^{1/d} \gg r_\omega$ (notice the constant density of states for particle–hole excitations) when $\omega < I_0 \ll A$ (d = dimension of the system). Thus, the majority of states that surround a given excited pair have energy of order $A \gg \omega$ and their occupations are only weakly affected by the creation of the particle–hole pair, which induces energy changes of order $e^2/a_0 \ll A$. Therefore, as a first approximation, one can replace the system by a dilute gas of randomly distributed particle–hole pairs interacting with each other only *via* the Coulomb potential (Shklovskii and Efros 1981) and with tunnelling effects between members of a pair only.

To describe a particle–hole pair, we consider the two-site Hamiltonian (Shklovskii and Efros 1981)

$$H_{1,2} = \psi_1 n_1 + \psi_2 n_2 + (e^2/r_{12}) n_1 n_2 + I(r_{12})(a_1^+ a_2 + a_2^+ a_1) \qquad (2)$$

which describes a particle–hole pair localized at sites $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$,

$$\psi_i = \varphi_i + \sum_{j \neq 1,2} (e^2/r_{ij}) n_j$$

and $I(r)$ is the overlap integral. The occupation numbers n_j ($j \neq 1, 2$) are assumed to stay unchanged during the excitation process (Shklovskii and Efros 1981). In the presence of the external potential $v(r) = Ze^2/r$, $\varphi_1 \to \varphi_1 + Ze^2/r_1$ and $\varphi_2 \to \varphi_2 + Ze^2/r_2$.

As in the 'classical' case ($I(r) = 0$), a very interesting property of the above Hamiltonian is the strong enhancement of low-energy densities of states for particle–hole excitations due to the Coulomb interaction. Let $|+\rangle$ and $|-\rangle$ denote the eigenstates of the two-site Hamiltonian (in the absence of the external potential) with $E_+ > E_-$ so that in the ground state, the state $|-\rangle$ is occupied and the state $|+\rangle$ is empty. Let $E_2 = E_+ + E_- + e^2/r_{12}$ denote the energy when both states are occupied. The probability of finding such a pair in the system is given by (Efros and Shklovskii 1975, 1981)

$$P(1,2) = F(\psi_1, \boldsymbol{r}_1; \psi_2, \boldsymbol{r}_2)\theta(\mu - E_-)\theta(E_2 - E_- - \mu) \tag{3}$$

where $F(\psi_1, \boldsymbol{r}_1; \psi_2, \boldsymbol{r}_2)$ is the probability of having on-site energies ψ_1 at $\boldsymbol{r}_1$ and ψ_2 at $\boldsymbol{r}_2$, and μ (we shall set $\mu = 0$ in the following) is the chemical potential. At energies $|\psi_1|, \psi_2| \gg \Delta$, where $\Delta \sim$ width of the Coulomb gap ($\Delta \sim A\gamma^{3/2}$(3D) and $A\gamma^2$(2D) (Efros and Shklovskii 1975, 1976) where $\gamma = (e^2/a_0)/A)$), F is essentially a constant, given by $F \sim (2Aa_0^d)^{-2}$. In this case, it can be shown using equation (3) that the density of states for a particle–hole excitation with energy ω is given by $\rho(\omega) \sim (\omega + e^2/r_{\omega)}$ (Shklovskii and Efros 1981), whereas for a non-interacting Fermi glass, $\rho(\omega) \sim \omega$. This estimate breaks down when $|\psi_1|, |\psi_2| < \Delta$, when the probability density F is reduced by the presence of the Coulomb gap (e.g. $F \sim \psi_1^2\psi_2^2$ in 3D), resulting in a weaker density of states $\rho(\omega) \sim (e^2/r_\omega)^{2d-1}$ and 2D and 3D. A straightforward analysis shows that the effect of the Coulomb gap is important only when $\omega \leqslant \omega_0$, where $e^2/r_{\omega_0} \sim \Delta$. Physically, in the presence of $I(r)$, the minimum excitation energy for a particle–hole pair separated by distance r is $I(r)$ (when $\psi_1 = \psi_2$). Thus for excitation energy ω, a pair is excited only when the distance between the two sites of the pair is of order $r > r_\omega$. At energies $\omega \ll \omega_0$, $r_\omega \gg e^2/\Delta$ and the Coulomb interaction between the pair is too weak to compensate for the Coulomb gap. Notice that in the 'classical' case $I(r) = 0$, the size of particle–hole pairs is independent of their energy (Efros and Shklovskii 1975, 1976). As a result, the above effect is absent and we always have $\rho(\omega)$ approximately constant as $\omega \to 0$.

Because of the much larger density of states, naively one might expect to find a stronger 'orthogonality catastrophe' effect in Coulomb glass systems than in metals, at least in the frequency range $\omega > \omega_0$. However, electronic states in a Coulomb glass are localized. Therefore, with a short-ranged potential, only a finite number of electrons can be affected, in contrast to the case of metals in which all electrons are involved. Thus one expects no 'orthogonality catastrophe' effect under short-ranged external potentials in Coulomb glass systems and the long-range nature of the potential is crucial. As a consequence, dimensional dependence of the final result is also expected.

Let $|+'\rangle$ and $|-'\rangle$ denote the corresponding eigenstates (cf $|+\rangle$ and $|-\rangle$) of the two-site Hamiltonian when the external potential $v(r)$ is present. It is straightforward to show that

$$|\langle + | -'\rangle|^2 = [I(r_{12})^2(\Delta_v + \Gamma' - \Gamma)^2/\Gamma\Gamma'(\Gamma' + \psi_2 - \psi_1 - \Delta_v)(\Gamma + \psi_2 - \psi_1)] \tag{4}$$

where

$$\Gamma = [(\psi_2 - \psi_1)^2 + 4I(r_{12})^2]^{1/2} \tag{5a}$$

$$\Gamma' = [(\psi_2 - \psi_1 + \Delta_v)^2 + 4I(r_{12})^2]^{1/2} \tag{5b}$$

$$\Delta_v = v(\boldsymbol{r}_2) - v(\boldsymbol{r}_1) = Ze^2/r_2 - Ze^2/r_1. \tag{5c}$$

Now, making the 'dilute particle–hole gas' approximation, and neglecting interactions between particle–hole pairs (we shall discuss the effect of screening later), our Coulomb glass system reduces essentially to a system of a non-interacting Fermi gas of localized states. Therefore, we can use equation (1) to estimate the overlap integral between the two ground states, where n denotes an empty localized state in the absence of an external potential, and n' denotes an occupied state in the presence of a potential, with $|A_{nn'}|^2$ given essentially by equation (4). We shall estimate the integral in the following.

First of all we distinguish between two kinds of pairs:

(i) those pairs in which the sign of $\psi_2 - \psi_1$ remains unchanged when an external potential is applied, i.e., $(\psi_2 - \psi_1)(\psi_2 - \psi_1 + v(\boldsymbol{r}_2) - v(\boldsymbol{r}_1)) > 0$, and

(ii) those pairs in which $\psi_2 - \psi_1$ changes sign.

In the absence of quantum mechanical tunnelling $I(r)$, only the second kind of pairs contribute where electrons move to new sites which are lower in energy in the external potential. This is just the polaron process discussed by Efros and Shklovskii (1975, 1976). In the present case, for weak external perturbations ($e^2/a_0 \ll A$), the first kind of pairs have typically small $|A_{nn'}|^2$ and their contribution can be estimated by their associated sum (Anderson 1967) $S = \Sigma_{nn'}|A_{nn'}|^2$, where $\langle G|G'\rangle \simeq e^{-S/2}$.

More interestingly (and usefully), we estimate the quantity

$$\tilde{S}(\omega) = \sum_{n,n'(E_n - E_{n'} > \omega)} |A_{nn'}|^2 \tag{6}$$

which determines the long-time behaviour of the Green function describing the removal (or addition) of an electron to the Coulomb glass system (Mahan 1974, 1981), $G(t) \sim \mathrm{e}^{-\tilde{S}(-1/it)}$ as $t \to \infty$. Fourier transforming, we obtain the spectral function of the Green function at small ω, which can be observed in, for example, a photo-emission experiment on deep-core electronic states.

Since $\tilde{S}$ involves sums over a large number of states randomly distributed in space, we shall estimate it by calculating its ensemble average, i.e.,

$$\tilde{S}(\omega) \simeq \int_{-A}^{A} \mathrm{d}\psi_1 \int_{\max(\psi_1, \psi_1 - \Delta_v)}^{A} \mathrm{d}\psi_2 \int \mathrm{d}\boldsymbol{r}_1 \int \mathrm{d}\boldsymbol{r}_2\, P(1,2)\, |\langle +|-'\rangle|^2_{r_1,r_2}\, \theta(E_+ - E_{-'} - \omega) \tag{7}$$

where $|\langle +|-'\rangle|^2$ is given by equation (4) and $P(1,2)$ is given by equation (3). Notice that the restriction to the first kind of pair is built into the limit of integration over ψ_2. Integral (7) can be estimated in a manner similar to that of the calculation of AC conductivity by Shklovskii and Efros (1981) in these systems. After some lengthy algebra, we find

$$\tilde{S}(\omega) \geqslant \frac{a_0}{(2Aa_0^d)^2} \int_{\omega}^{2I_0} \mathrm{d}\Omega \left[r\Omega^{d-1} \rho(\Omega) \int_{r\Omega}^{\infty} R^{d-1}\, \mathrm{d}R\, |A(\Omega, R)|^2 \right] \tag{8a}$$

where

$$|A(\Omega, R)|^2 = (Ze^2 r\Omega/R^2)^2/(\Omega + (Ze^2 r\Omega/R^2))^2. \tag{8b}$$

$\rho(\omega)$ is the density of states for particle–hole excitation. $|A(\omega, R)|^2 \sim |A_{nn'}|^2$ when the two sites of the pair are located at a distance r_ω from each other, and at a distance $R \gg r_\omega$

from the external added charge, giving $\Delta_v \sim v(\boldsymbol{R} + \boldsymbol{r}_\omega) - v(\boldsymbol{R}) \sim Ze^2 r_\omega/R^2$. Performing the integral, we obtain for $\omega > \omega_0$,

$$\tilde{S}(\omega) \geqslant \alpha Z^{3/2}\gamma^2[\ln(2I_0/\omega)]^{5/2}((e^2/a_0)/\omega)^{1/2} \qquad \text{(3D)} \qquad (9a)$$

$$\geqslant \alpha' Z\gamma^2[\ln(2I_0/\omega)]^2 \qquad \text{(2D)} \qquad (9b)$$

$$\sim \text{constant} \qquad \text{(1D)} \qquad (9c)$$

and, for $\omega \leqslant \omega_0$,

$$\tilde{S}(\omega) \geqslant \alpha Z^{3/2}[\ln(2I_0/\omega)]^{-3/2}((e^2/a_0)/\omega)^{1/2} \qquad \text{(3D)} \qquad (10a)$$

$$\sim Z\ln(\ln\omega) \qquad \text{(2D)} \qquad (10b)$$

$$\sim \text{constant} \qquad \text{(1D)} \qquad (10c)$$

where α and α' are constants of order 10^{-1}.

The effect of screening can be included by replacing $v(r)$ by $v_{\text{sr}}(r, \omega)$ where $v_{\text{sr}}(q, \omega) = v(q)/\varepsilon(q, \omega)$, $\varepsilon(q, \omega)$ is the dielectric function. In the limit $q \to 0$, $\varepsilon(q, \omega) \to 1 + b(\omega)$, where in 3D, $b(\omega) \sim e^4 r_\omega^4/(2Aa_0^3)^2 \ll 1$ for $\omega > \omega_0$ and $b(\omega) \sim \ln(\ln\omega)$ for $\omega < \omega_0$ (Bhatt and Ramakrishman 1984). Since in 3D, the orthogonal catastrophe effect is dominated by the factor $((e^2/a_0)/\omega)^{1/2}$, the additional ln ω term is not going to change the result qualitatively. In 2D, however, a similar analysis indicates that although $\tilde{S}(\omega)$ remains unchanged for $\omega > \omega_0$, the $Z\ln(\ln\omega)$ term is destroyed by screening at $\omega < \omega_0$, indicating that there is no true orthogonality catastrophe in 2D. The physical reason for weak screening is that within the two-site Hamiltonian, the system can be considered as a dilute gas of localized particle–hole pairs, where isolated charges are rare (Coulomb gap). Thus instead of thinking of the system as a collection of point charges, one should think of it as a collection of dipoles. The interaction between dipoles is weak, which results in the observed weak screening.

There are several interesting features worth noticing in these results. First of all, we predict a true orthogonality catastrophe effect only in 3D ($e^{-S} \to 0$). Secondly, the strong enhancement in the density of states $\rho(\omega)$ and the long-rangeness of the potential are both crucial in the present problem. In fact, it is easy to show from equation (8) that there exists no orthogonality catastrophe in any dimension if $\rho(\omega) \sim \omega$ or if we let $A(\Omega, R) = 0$ for $R > R_c$ (R_c = range of the potential). Another interesting result is that S is not proportional to Z^2 for small Z, so that there exists *no linear response regime.* This is a direct effect of the long-range potential, as can be seen by evaluating integral (8). The difference in $\tilde{S}(\omega)$ for ω above and below ω_0 can be understood quite easily. The inclusion of the Coulomb gap introduces factors of order $[\ln(2I_0/\omega)]^{-(2d-1)}$ into $\rho(\omega)$. In 3D, the orthogonal catastrophe effect is dominated by the factor $((e^2/a_0)/\omega)^{1/2}$, and is not affected qualitatively by the inclusion of additional logarithmic terms. In 2D, however, the orthogonal catastrophe effect itself exhibits logarithmic divergence. Thus the presence of the Coulomb gap has a strong effect on its behaviour.

The second kind of pair is a generalization of the polaron process (Efros and Shklovskii 1975, 1976) to the case for $I(r) \neq 0$. They have typically large $A_{nn'}$ and thus contribute a factor of order δ^M, where δ is a number of order 10^{-1} and M is the number of such pairs found in the system. The number M can be estimated in a manner similar to that of the estimation of the polaron gap by Efros (1976). We find that their contribution is similar in magnitude to that from the first kind of pair except that it is smaller by a factor of order $\ln(2I_0/\omega)$ (2D and 3D). Thus the qualitative results obtained from process (i) remain unaffected.

Experimentally, our results indicate that the orthogonality catastrophe effect will be observable in 3D and 2D systems, with qualitatively different behaviours. In a photo-emission experiment on a deep core electronic state with energy E_0 (see e.g. Hollinger *et al* 1985), the sharp emission line will be broadened *asymmetrically*. In 2D, a power law type behaviour for the spectral function will be found in the frequency range $\omega \geqslant \omega_0 + E_0$, saturating at $\omega \sim \omega_0 + E_0$, and leaves a δ-function peak with reduced weight at $\omega \sim E_0$. In 3D, the δ-function peak in the spectral function at $\omega = E_0$ will be completely suppressed, resulting in a finite, broad spectral function distributed at the frequency range $E_0 + \min(2I_0, e^2/a_0) \geqslant \omega \geqslant E_0$ which is asymmetric ($A(\omega) = 0$ for $\omega < E_0$) instead of a simple Lorentzian lineshape. The orthogonality catastrophe effect also leads to a further broadening of the Coulomb + polaron gap in the one-particle density of states. However this effect is difficult to observe experimentally.

We would like to make a few final remarks here. To simplify the study, we have introduced the dilute particle–hole gas approximation for a Coulomb glass system neglecting the interaction between particle–hole pairs. The validity of these approximations is based on physical arguments which unfortunately are hard to justify rigorously. Another crucial approximation in our analysis is the replacement of $\tilde{S}$ by its average. To test this approximation, we have evaluated the mean square fluctuation $\langle \tilde{S}^2 \rangle = \langle (\tilde{S} - \langle \tilde{S} \rangle)^2 \rangle$ and have found that $\langle \tilde{S}^2 \rangle \leqslant \langle \tilde{S} \rangle$ to the leading $1/\omega$ dependence, suggesting that the mean $\langle \tilde{S} \rangle$ is a good approximation for $\tilde{S}$ in our study.

We find that a little quantum mechanical tunnelling, in combination with Coulomb interaction, leads to an orthogonality catastrophe type of effect in Coulomb glass systems. This effect can be observed in photo-emission experiments on deep core electronic states, which would provide us with an alternative way of confirming the theoretically predicted Coulomb correlation in Coulomb glass materials. (This effect is absent in a non-interacting Fermi glass, see discussion after equation (9).) This is the main result of the paper.

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References

Anderson P W 1967 *Phys. Rev. Lett.* **18** 1049
Baranovskii S D, Shklovskii B I and Efros A L 1980 *Zh. Eksp. Teor. Fiz.* **78** 395 (Engl. Transl. *Sov. Phys.–JETP* **51** 199)
Bhatt R N and Ramakrishman T V 1984 *J. Phys. C: Solid State Phys.* **17** L639
Davies J H, Lee P A and Rice T M 1984 *Phys. Rev.* B **29** 4260
Efros A L and Shklovskii B I 1975 *J. Phys. C: Solid State Phys.* **8** L49
Efros A L 1976 *J. Phys. C: Solid State Phys.* **9** 2021
Efros A L and Shklovskii B I 1985 *Electron–Electron Interactions in Disordered Systems* ed A L Efros and M Pollak (Amsterdam: North Holland)
Hollinger G *et al* 1985 *Phys. Rev.* B **32** 1987
Mahan G D 1974 *Solid State Phys.* **29** ed H Ehrenreich, F Scitz and D Turnbull (New York: Academic Press)
Mahan G D 1981 *Many-Particle Physics* (New York: Plenum)
Pollak M and Ortunö M 1985 *Electron–Electron Interactions in Disordered Systems* ed A L Efros and M Pollak (Amsterdam: North Holland)
Shklovskii B I and Efros A L 1981 *Zh. Eksp. Teor. Fiz.* **81** 406 (Engl. Transl. *Sov. Phys.–JETP* **54** 218)