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Dielectric Response, Quantum Geometry, and Bounds on Optical Gaps

1. **The quantum geometric origin of capacitance in insulators**
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   Nat. Commun. **15**, 4621 (2024); https://doi.org/10.1038/s41467-024-48808-x
   https://doi.org/10.48550/arXiv.2306.08035

2. **Universal relation between energy gap and dielectric constant**
   Authors: Yugo Onishi and Liang Fu
   https://doi.org/10.48550/arXiv.2401.04180

Recommended with a Commentary by Steven M. Girvin (Yale Quantum Institute) & Kun Yang (Florida State University)

1 Introduction

It is well understood that for a harmonic oscillator of mass m, increasing the spring stiffness k lowers the susceptibility χ of the oscillator to displacement by an external force F

$$\langle x \rangle = \chi F = \frac{1}{k} F, \tag{1}$$

and raises the (quantum) excitation gap $\hbar\omega = \hbar\sqrt{\frac{k}{m}}$. This simple example provides the intuition behind the fact that solids with large bandgaps tend to have smaller dielectric constants. This relationship has been explored in the past [1] but Komissarov, Holder, and Queiroz bring a modern view in terms of the theory of polarization relating it to the quantum geometry of the electronic wave functions of insulators. They demonstrate that the electronic contribution to the capacitance is quantized for a 2D electron gas in a strong magnetic field and present an interesting symmetry argument for why diamond is an outlier with a large index of refraction despite its large bandgap.

Onishi and Fu provide a rigorous derivation of an inequality connecting the bandgap and the dielectric constant and note some materials that come close to saturating the inequality. Onishi and Fu have also recently explored the relationship between the negative first frequency moment of the optical conductivity and the static structure factor in terms of the ground-state Chern number for topological materials [2] and demonstrated a fundamental bound on the energy gap of topological insulators [3].

Both of the recommended papers present modern data compilations including 2D van der Waals materials.

2 Simple example of f-sum rule and gap bound

Here we illustrate the oscillator strength (f-sum) rule by deriving it in the simplest setting, namely a single electron system in 1D, using elementary methods. As we will see, the sum rule is intimately connected to charge conservation and the associated gauge invariance [4].

Consider the one-body Hamiltonian

$$H_0 = \frac{p^2}{2m} + V(x), \tag{2}$$

where $V(x \to \pm\infty) = \infty$ so that the electron is bound and has a discrete spectrum. Charge conservation licenses us to consider a gauge transformed version of the Hamiltonian with (for simplicity) a constant vector potential $A(x) = A = \partial_x(Ax)$

$$H(A) = \frac{(p + eA)^2}{2m} + V(x) = H_0 + \Delta H(A), \tag{3}$$

where

$$\Delta H(A) = eAv + \frac{e^2A^2}{2m}. \tag{4}$$

In the above $v = p/m$ is the velocity operator when $A = 0$. A constant vector potential is a pure gauge, so the spectra of H_0 and $H(A)$ are identical. Another way to see this simple fact is to note that if $\psi_j(x)$ is an eigenfunction of H_0 with eigenvalue ϵ_j, then

$$\psi'(x) = e^{-ieAx}\psi(x) \tag{5}$$

is an eigenstate of $H(A)$ with the same eigenvalue. Nevertheless we can treat $\Delta H(A)$ as a perturbation, and derive non-trivial results using the fact that energy levels actually do *not* change [4].

Since the average velocity in a bound state must be zero, the first term of Eq. (4) only contributes at second-order in perturbation in A. The second term in Eq. (4) is a constant energy shift which is also second-order in A. Combining the effects of these two terms yields

$$\Delta\epsilon_n^{(2)} = e^2A^2 \sum_{l\neq n} \frac{|\langle l|v|n\rangle|^2}{\epsilon_n - \epsilon_l} + \frac{e^2A^2}{2m} = \frac{e^2A^2}{\hbar^2} \sum_{l\neq n} \frac{\langle l|[x, H]|n\rangle|^2}{\epsilon_n - \epsilon_l} + \frac{e^2A^2}{2m} \tag{6}$$

$$= \frac{e^2A^2}{\hbar^2} \sum_{l\neq n} |\langle l|x|n\rangle|^2(\epsilon_n - \epsilon_l) + \frac{e^2A^2}{2m} = 0, \tag{7}$$

from which we obtain the so-called oscillator strength sum rule for dipole transitions out of level n

$$\sum_{l\neq n} |\langle l|x|n\rangle|^2(\epsilon_l - \epsilon_n) = \frac{\hbar^2}{2m}. \tag{8}$$

The dimensionless oscillator strength for the transition from state n to state l is defined to be

$$f_{nl} = \frac{2m}{\hbar^2}\langle l|x|n\rangle|^2(\epsilon_l - \epsilon_n), \tag{9}$$

so that the f-sum rule becomes

$$\sum_{l\neq n} f_{nl} = 1. \tag{10}$$

Note that for a harmonic oscillator in its ground state, the sum rule is saturated by the transition from $n = 0$ to $l = 1$ since $x|0\rangle$ is an exact (unnormalized) eigenstate of the Hamiltonian. Thus f_{0l} is a measure of the strength of optical absorption of the atom relative to that of a harmonic oscillator with with frequency $\hbar\omega_{0l} = \epsilon_l - \epsilon_0$. Furthermore, the matrix elements $\langle n \pm 1|x|n\rangle$ are such that the same result is obtained for any initial state, not just the ground state.

The connection with charge conservation is even clearer for the case of multi-electron atoms containing N electrons, for which an additional factor of N appears on the RHS of this equation. The sum over states (and the frequency integral discussed below) has to be carried out to high excitation levels to saturate the sum rule by capturing excitations of tightly bound electrons as well as all the many-electron ('shake-up') transitions induced by Coulomb correlations among the electrons. Because in some atoms (e.g., the alkali atoms) there is a substantial gap between the hydrogen-like single valence electron excitations and the single core electron (and many-electron) excitations, truncating the sum over excited state energies below this higher gap gives an approximate f-sum corresponding to $N \sim 1$. Ref. [5] explores the issue of f-sums with finite energy cutoffs for valence electrons in solids.

Below we will use this sum rule and the static susceptibility to find a bound on the excitation gap of solids. First however, we will take a short detour to relate the sum rule to an integral of the conductivity over frequency by the following argument. Consider a (seemingly very different) problem, namely perturbing the system initially in state $|n\rangle$ with a uniform but possibly time-dependent electric field $E(t)$, so

$$\Delta H(E) = eEx, \tag{11}$$

and use Fermi's Golden rule to calculate the energy absorption rate when $E(t) = E_0 e^{-i\omega t}$:

$$P(\omega) = \frac{2\pi}{\hbar} e^2 E_0^2 \sum_{l\neq n} |\langle l|x|n\rangle|^2 (\epsilon_l - \epsilon_n)\delta(\epsilon_l - \epsilon_n - \hbar\omega) = \sigma^{\rm abs}(\omega) E_0^2, \tag{12}$$

where $\sigma^{\rm abs}(\omega)$ is the real or absorption part of the frequency-dependent conductivity. Combining with (8), we find

$$\int_{-\infty}^{+\infty} \sigma^{\rm abs}(\omega) d\omega = \frac{\pi e^2}{m}, \tag{13}$$

which is equivalent to the single-electron version of the f-sum rule, Eq. (3) of Onishi and Fu.

Returning from our detour, we now consider the even simpler case of $\omega = 0$, namely E is time-independent, and consider its effect on ground ($n = 0$) state energy to 2nd order:

$$\Delta\epsilon_0^{(2)} = e^2 E^2 \sum_{l\neq 0} \frac{|\langle l|x|0\rangle|^2}{\epsilon_0 - \epsilon_l} = -\chi^0 E^2/2, \tag{14}$$

where the 2nd equation defines the static polarizability

$$\chi^0 = 2e^2 \sum_{l\neq 0} \frac{|\langle l|x|0\rangle|^2}{\epsilon_l - \epsilon_0} = 2e^2 \sum_{l\neq 0} \frac{|\langle l|x|0\rangle|^2(\epsilon_l - \epsilon_0)}{(\epsilon_l - \epsilon_0)^2} \quad (15)$$

$$\leq \frac{e^2}{\Delta^2} \sum_{l\neq 0} |\langle l|x|0\rangle|^2(\epsilon_l - \epsilon_0) = \frac{\hbar^2 e^2}{m\Delta^2}, \quad (16)$$

where $\Delta = \epsilon_1 - \epsilon_0$ is the energy gap, and we used the sum rule (8) in the last step. We thus find

$$\Delta \leq \hbar e/\sqrt{m\chi^0}, \quad (17)$$

which is the one-electron version of Eq. (10) of Onishi and Fu.

Recall that the single-mode-approximation assumes that $x|0\rangle$ is an eigenstate and therefore saturates the sum rule. If this is the case then the inequality on the gap becomes an equality. This is of course the case for a harmonic oscillator. Onishi and Fu present an interesting compilation of data on the bandgap/dielectric constant correlation for a wide variety of materials including novel 2D systems such as cubic boron nitride (c-BN) which comes close to saturating the bound for the (transverse) excitation gap.

3 Tranverse vs. longitudinal dielectric response

A slightly confusing point in electrodynamics discussed by Onishi and Fu is the difference in physics represented by the poles and zeros of the dielectric function $\epsilon(\omega)$, the latter corresponding to poles of the inverse dielectric function $\frac{1}{\epsilon(\omega)}$. Examination of Maxwell's equations shows that the poles of $\epsilon(\omega)$ determine that transverse electromagnetic mode frequencies, whereas the poles of $\frac{1}{\epsilon(\omega)}$ determine the longitudinal mode frequencies (i.e., the plasmons), the difference being related to the fact that longitudinal external perturbations induce screening electric fields which modify the self-consistent response.

References

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