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Preparatory School to the Winter College on Optics and the Winter College on Optics: Advances in Nano-Optics and Plasmonics

30 January - 17 February, 2012

Emission and scattering of light in nanostructures and ways to modify probabilities of quantum transitions

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- Introduction
- Historical comments
- DOS and uncertainty relation
- DOS in statistical physics
- DOS and probability of quantum transitions
- DOS in optics: emission and scattering of light
- Integral and local density of states





Introduction:

A first attempt to clarify what is DOS



DOS for different scales

 $D(p)dp = D(E)dE = D(\lambda)d\lambda = D(\omega)d\omega = D(k)dk,$

$$D(p) = D(k)\frac{dk}{dp}; D(\omega) = D(k)\frac{dk}{d\omega}; D(\lambda) = \frac{dk}{d\lambda}, D(E) = D(k)\frac{dk}{dE}$$

The result for DOS is independent of shape of a volume chosen for calculations: "It is not possible to hear the shape of a drum" (H. Weyl, 1930-ies)

Schroeder M Fractals, Chaos, Power Laws (NY: Freeman and Co, 1990)

DOS for m = 0 and $m \neq 0$

$$D_3(k) = \frac{k^2}{2\pi^2}$$
. + $D(E) = D(k)\frac{dk}{dE}$

Take dispersion relation w(k) and E(p)

$$\omega = ck$$

$$E = \frac{p^2}{2m} = \frac{\hbar^2 k^2}{2m}$$

-

and arrive at

$$D_{3\gamma}(\omega) = \frac{\omega^2 n^3}{2\pi^2 c^3}$$

$$D_{3e}(E) = \frac{m^{3/2} E^{1/2}}{2^{1/2} \pi^2 \hbar^3}$$

EM-waves DOS for photons and electrons in 3d-space



Historical comments

"The physicist prefers to forget the way which led him to discovery" P. A. M. Dirac, 1972



Lord Rayleigh *Philos. Mag.* **49** 539 (1900)



John Strutt (lord Rayleigh) 1842 – 1919

- Rayleigh was the first to propose counting of standing EM-waves in a 3dcavity to get formula for equilibrium EM-radiation (black body radiation)
- It is now referred to as the <u>Rayleigh-Jeans formula</u>

Planck M Ann. Phys. 1 69 (1900)



Max Planck (1858 - 1947)

$$U(\omega) = \frac{\hbar\omega^3}{\pi^2 c^3} \frac{1}{\exp(\hbar\omega/k_B T) - 1}$$

Planck did not count modes, instead he considered energy emitted by a classical cavity and found it proportional to w^2/c^3

Only in 1924 Bose extended the Rayleigh's approach to get this formula in terms of statistical physics

What happened between the famous work by Rayleigh in <u>1900</u> and the ingenious paper by S. N. Bose in <u>1924</u>?



Lui de Broglie 1892-1987

1906 Planck proposed to consider phase space in terms of elementary cells =h

1922 de Broglie took the Planck's idea and derived the Rayleigh -Jeans formula (J. de Phys. Nov. 1922)

1924 de Broglie applied for the first time the concept of DOS not to EM-waves, but to atoms, i.e. to the particles of matter (Phil. Mag. and J. Sci. 47 446, 1924)

DOS for quantum particles D(p) then is simply the number of cells h in the phase space interval **dpdx for 1d space**. In 3d DOS is the number of cells h^3 in a layer $4\pi p^2 dpdxdydz$

N. Bose: The beginning of quantum statistic



Equilibrium EM-radiation = gas of photons

$$N(\omega,T) = D(\omega)F(\omega,T)d\omega$$

$$U(\boldsymbol{\omega}) = E(\boldsymbol{\omega})N(\boldsymbol{\omega},T)$$

$$F(\omega) = [\exp(\hbar\omega/k_B T) - 1]^{-1}$$

$$U(\omega)d\omega = \frac{\hbar\omega^3}{\pi^2 c^3} \frac{1}{\exp(\hbar\omega/k_B T) - 1} d\omega$$

Bose S. N. *Z. Physik* **26** 181,1924

Shatiendranat Bose (1894-1974)

Extention of Bose's approach to atoms (bosons) and electrons (fermions)

1924 application of DOS to statistic of atoms (Bose-Einstein statistic) Einstein A Sitz. Preuss. Akad. Wiss. 22 261

(1924)

Fermi E Z. Physik 36 902 (1926)

 1927 application of DOS to statistic of electrons (Fermi - Dirac statistic)
Dirac P A M Proc. Royal Soc. London 112 661 (1926)
Fermi E Rend Lincei 6 602 (1927).

DOS and the uncertainty relation

Take $\Delta k=\pi/L$, i.e. $\Delta kL=\pi$ (Rayleigh 1900) Multiple both parts $h/2\pi$ to get $h\Delta kL/2\pi=h/2$ Remember $p=(h/2\pi)k$, i.e. $\Delta p=(h/2\pi)\Delta k$ (de Broglie 1923) Consider L= Δx and arrive at $\Delta p\Delta x=h/2$

Uncertainty relation comes immediately from mode discreteness in a cavity and ascribing wave properties to particles of matter.

It was contained in 1906 Planck's idea of the elementary cell h in phase space: For quantum particles phase space is discrete and consists of elementary cells h.

Number of cells per unit volume -> number of states.

Then DOS represents the number of states contained in a unit p interval, i.e. dN(p)=D(p)dp 16

W. Heisenberg clearly understood that he follows the Planck's idea.

He wrote in his paper:

"The uncertainty relation expresses explicitly the facts which have been treated previously in terms of breaking the phase space into elementary h cells"

W. Heisenberg "Über den anschaulichen Inhalt der quantentheoretischen Kinematik und Mekhanik" Z. Phys. 43, 172, 1927 ("On the visual content of quantum-theoretical kinematics and mechanics") DOS in statistical physics Radiation statistic (Bose 1924)

$$N(\omega,T) = D(\omega)F(\omega,T)d\omega$$

<u>Particle statistic</u> (atoms, molecules in gases, electron gas in semiconductors)



$$N_{E_1,E_2} = \int_{E_1}^{E_2} f(E,T) D(E) dE$$

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Erwin Shroedinger did recognized great heuristic value of the DOS concept.

E.Shroedinger 1897-1961



it is very instructive to compare density of particles (atoms, molecules, electrons) with the density of states. If particle density in space is <u>small</u> as compared to DOS we can apply <u>classical physics</u>. If particle density is noticeable as compared to DOS or even <u>approaches DOS value</u>, then only <u>quantum approach</u> is adequate.

DOS and probability of quantum transitions

What is the "Fermi Golden Rule"?

If a quantum system experiences a transition from initial state to the final state belonging to a continuum of states then the probability of such a transition is proportional to the density of final states.

$$W = \frac{2\pi}{\hbar} \left| H \right|^2 D(E)$$

This statement is derived based on QM perturbation theory. The rate of transition is proportional to the number of ways in which the transition can be performed. This statement plays a big role in the theory of elementary particles collisions and scattering. It is for this reason Fermi called it "The Golden rule".

<u>D(E) is the particle DOS.</u> By no means spontaneous emission can be derived on that basis. The formula can be used if EM-field is considered as perturbation to calculate STIMULATED absorption and emission of photons but again DOS means number of particle states. E.g. in a semiconductor for absorption these are free electron states in the c-band, for emission these are hole states available in the v-band

Fermi did it in Nuclear Physics (Chicago: Univ. Chicago Press, 1950)



Enrico Fermi (1901 - 1954)

Schiff L I Quantum Mechanics (NY .: McGraw-Hill Book Company, 1949, 1955);

Page 141-142

Returning to the nuclear reaction $A + a \longrightarrow B + b$, we use a general principle of quantum mechanics to derive some essentially statistical results on the variation of the cross-section. From quantum mechanics, the probability per unit time of

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Ch. VIII

142 Nuclear Reactions transition = number of transitions per unit time = w is given by "Golden Rule No. 2": *

$$w = \frac{2\pi}{h} \left| \right|_{\ell} \left| \frac{2 dn}{dE} \right| \qquad \text{VIII.2}$$

where H is the matrix element of the perturbation causing the transition, and dn/dE = energy density of final states, counting each degenerate state separately.

* Derived in Schiff, Quantum Mechanics, p. 193. ("Golden Rule No. 1" is on page 148 of this text).

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Compound Nucleus

Ch. VIII

The explanation of this phenomenon is based on the assumption that the transition $A + a \rightarrow B + b$ occurs through an intermediate state C:

$$A + a \longrightarrow C \longrightarrow B + b$$

State C is the "Compound nucleus". The idea of the compound nucleus is due to Bohr.*

The idea of how resonances in cross section result from this assumption can be obtained from the quantum mechanics of second order transitions. The probability of transition, per unit time. is given by "Golden Rule No. 1":**

trans. prob./sec =
$$\frac{2\pi}{K} \left| \frac{\mathcal{H}_{cA}}{E_{A} - E_{c}} \right|^{2} \left| \frac{\text{density of}}{\text{states}} \right|$$
 VIII.19

provided there are no direct transitions from A to B. The cross

* Bohr, <u>Nature 137</u> 344 (1936) ** Schiff, p. 196, eq. (29.20)

Fermi ascribed notation "Golden rule" for QM-perturbational formula.

He never claimed he derived this formula.

He never ascribed this notation to spontaneous optical transitions probability which is essentially a QED-problem.

Therefore the very notation "Fermi golden rule" in quantum mechanics is not justified at all. It is by no means can be applied to spontaneous transitions rates in QED.

This confusing situation gives rise to many discussions, e.g. in Phys. Today, Aug. 2001

DOS in optics: Spontaneous emission and scattering of light



In 1916 in his pioneering paper Einstein introduced stimulated and spontaneous emission rates and derived Planck's formula for black body radiation in terms of balanced transitions between discrete energy levels.

He found it is necessary to assume

$$A / B = \frac{\hbar \omega^3}{\pi^2 c^3}$$

Albert Einstein 1879-1955

Verh. Deutsch. Phys. Ges. 18 318 (1916) Could Einstein formulate the so-called "Fermi Golden Rule" in 1916? Yes, he could. But he didn't. Based on the Rayleigh's approach of counting modes and Planck's E=hv, Einstein in 1916 (!) could make the statement: "A is proportional to photon energy and number of EM-modes (photon states)" Instead only in 1927 Dirac made this statement 25

1927 - the beginning of Quantum Electrodynamics



Dirac quantized EM-field and introduced DOS of EM-field into spontaneous emission rate

the process. The probability of the absorption process is thus proportional to N_r' . In the same way the probability of a light-quantum in state r being emitted is proportional to $(N_r' + 1)$, and the probability of a light-quantum in state r being scattered into state s is proportional to $N_r'(N_s' + 1)$. Radiative **The Quantum Theory of the Emission and Absorption of Radiation**



P. A. M. Dirac

Proceedings of the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character, Vol. 114, No. 767 (Mar. 1, 1927), 243-265.

To establish a connection between the number of light-quanta per stationary state and the intensity of the radiation, we consider an enclosure of finite volume, A say, containing the radiation. The number of stationary states for light-quanta of a given type of polarisation whose frequency lies in the

262 P. A. M. Dirac.

range ν_r to $\nu_r + d\nu_r$ and whose direction of motion lies in the solid angle $d\omega_r$ about the direction of motion for state r will now be $A\nu_r^2 d\nu_r d\omega_r/c^3$. The energy of the light-quanta in these stationary states is thus $N_r' \cdot 2\pi h\nu_r \cdot A\nu_r^2 d\nu_r d\omega_r/c^3$. This must equal $Ac^{-1}I_r d\nu_r d\omega_r$, where I_r is the intensity per unit frequency range of the radiation about the state r. Hence

$$\mathbf{I}_r = \mathbf{N}_r' (2\pi\hbar) \nu_r^3 / c^2, \tag{28}$$

so that N_r' is proportional to I_r and $(N_r' + 1)$ is proportional to $I_r + (2\pi h)\nu_r^3/c^2$. We thus obtain that the probability of an absorption process is proportional to I_r , the incident intensity per unit frequency range, and that of an emission process is proportional to $I_r + (2\pi h)\nu_r^3/c^2$, which are just Einstein's laws.*





Do you feel the Dirac's spirit here?

QUANTUM THEORY OF RADIATION*

BY ENRICO FERMI UNIVERSITY OF ROME, ITALY

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For these reasons it is often more convenient to represent the field in the following way. Instead of considering the radiation in infinite space, let us consider the radiation enclosed in a cavity of finite volume Ω with perfectly reflecting walls. If afterwards we let the cavity become infinite in every direction, we shall get as a limit the properties of radiation in free space.

The electromagnetic vibrations in a cavity of finite volume, just as the vibrations of an elastic body of finite volume, may be represented by the superposition of a discreet infinity of fundamental vibrations each one corresponding to a system of standing waves. The number of standing vibrations whose frequency lies between ν and $\nu + d\nu$ is given, for a very large volume Ω , by:

$$dN = \frac{8\pi}{c^3} \,\Omega \nu^2 d\nu \tag{1}$$

c being the velocity of light. \lfloor

QUANTUM THEORY OF RADIATION

$$\gamma = \frac{16\pi^3 e^2}{\Omega h} \sum_{s} \frac{\nu^2}{\nu_s} (A_s X)^2 \sin^2 \Gamma_s \frac{1 - e^{\{\gamma - 2\pi i (\nu_s - \nu)\}t}}{-2\pi i (\nu - \nu_s) - \gamma}$$

The sum may be calculated by the following method: since the phase, direction and polarization of the different radiation components are distributed at random, we may substitute for $(A_sX)^2\sin^2\Gamma_s$ its mean value, taken over all phases, directions and polarizations; we replace then the sum by an integral over ν_s , multiplying by the factor:

$$\frac{8\pi}{c^3}\Omega\nu_s^2d\nu_s\tag{54}$$

which gives, according to (1) the number of radiation components with frequency between ν_s and $\nu_s + d\nu_s$. We get then, observing

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1946 - E. M. Purcell predicts modification of spontaneous emission rates in complex media



Edward M. Purcell b. 1912

Phys. Rev. 69, 681 (1946)

B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. PURCELL, Harvard University.—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

 $A_{\nu} = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2)$ sec.⁻¹,

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu = 10^7$ sec.⁻¹, $\mu = 1$ nuclear magneton, the corresponding relaxation time would be 5×1021 seconds! However, for a system coupled to a resonant electrical circuit, the factor $8\pi\nu^2/c^3$ no longer gives correctly the number of radiation oscillators per unit volume, in unit frequency range, there being now one oscillator in the frequency range ν/Q associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor $f = 3Q\lambda^3/4\pi^2 V$, where V is the volume of the resonator. If a is a dimension characteristic of the circuit so that $V \sim a^3$, and if δ is the skin-depth at frequency ν , $f \sim \lambda^3/a^2 \delta$. For a non-resonant circuit $f \sim \lambda^3/a^3$, and for $a < \delta$ it can be shown that $f \sim \lambda^3/a\delta^2$. If small metallic particles, of diameter 10-3 cm are mixed with a nuclear-magnetic medium at room temperature, spontaneous emission should establish thermal equilibrium in a time of the order of minutes, for $\nu = 10^7$ sec.⁻¹.

Modified DOS in mesoscopic structures due to confinement of EM-waves is generally recognized as basic physical phenomenon

Model mesoscopic structures in which this effect is being studied since 1970-ies:

- Media with n>1
- Thin layers
- Metal-dielectric interfaces
- Microcavities
- Photonic crystals (3d and 2d)
- Cell membranes
 - Nuclear reactions

Fluorescent probes: molecules, ions, Q-dots

An atom in front of a mirror



K. H. Drexhage. Influence of a dielectric interface on fluorescence decay time. *J. Luminescence*, **1–2** (1970), 693–701.

Nanostructures with modified DOS





Microcavities: spontaneous emission Purcell 1946 $\gamma = \frac{3}{4\pi^2} \frac{\lambda^3}{V} Q \gamma_0$

Bunkin and Oraevsky 1959

$$\gamma \sim \frac{Q}{V} \frac{\omega_c^2}{\omega_c^2 + Q^2 (\omega_c - \omega_a)^2}$$

Woggon and coworkers, 2001



Photonic crystals

Photonic crystal possess discontinuous DOS. This not only promise freezing of excited atomic/molecule states but make us consider the decay process as essentially non-Markovian, i.e. probability can not be used to describe it correctly (number of events is not proportional to time of detection).

First papers

Bykov V.P. 1972, Ohtaka 1979, Yablonovitch 1987, John 1987

Books

Joannopoulos J D, Meade R D, Winn J N Photonic Crystals: Molding theFlow of Light (Princeton 1995) Sakoda K Optical Properties of Photonic Crystals (Berlin 2001)



Lambropoulos et al. Rep. Prog. Phys. 63 455 (2000)

VOLUME 88, NUMBER 9

PHYSICAL REVIEW LETTERS

4 March 2002

Decay Distribution of Spontaneous Emission from an Assembly of Atoms in Photonic Crystals with Pseudogaps

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PHYSICAL REVIEW A, VOLUME 62, 013805

Spontaneous emission from a two-level atom in a three-dimensional photonic crystal

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DOS effects on resonant and Raman scattering

Elastic scattering (resonant, Rayleigh scattering) is also proportional to DOS Blue color of the sky $I(w): w^4(Rayleigh)$ includes DOS component ω^2



Anisotropic Light Scattering in Nanoporous Materials: A Photon Density of States Effect

NANO LETTERS

2004 Vol. 4, No. 9

1755-1758

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The DOS effect results in predictable and controllable modification of scattering and can be purposefully used in certain 42 light harvesting and illumination systems.

Inelastic scattering (nonresonant, Raman scattering)

$$W(\omega',\omega) = \frac{2\pi^2}{\hbar^2} \omega' \omega N(\omega') |S|^2 D(\omega)$$

SERS contains contribution from DOS Gaponenko S V *Phys. Rev.* B **65**, 140303 (R) (2002) Zuev V.S., Franzesson A.V., *Opt. Spektr.* **93**, 117 (2002)

PHYSICAL REVIEW B, VOLUME 65, 140303(R)

Effects of photon density of states on Raman scattering in mesoscopic structures

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Photon density of states (DOS) effects on spontaneous Raman scattering in certain mesoscopic structures are outlined. Similar to spontaneous emission spontaneous Raman scattering should experience modification if DOS redistribution over frequency and solid angle occurs. In continuous media DOS dependence on dimensionality d=3,2,1 results in different power dependencies of scattering rate on refraction index *n* and frequency ω in the form $(n\omega)^d$. Photon DOS effects can be purposefully used to enhance Raman signals in molecular spectroscopy using purely dielectric structures.

Understanding DOS via optical tunelling

VOLUME 88, NUMBER 9

PHYSICAL REVIEW LETTERS

4 March 2002

Imaging the Local Density of States of Optical Corrals

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Integral and Local DOS

Barnett-Loudon sum rule

VOLUME 77, NUMBER 12

PHYSICAL REVIEW LETTERS

16 September 1996

Sum Rule for Modified Spontaneous Emission Rates

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Rodney Loudon Department of Physics, University of Essex, Colchester C04 3SQ, England (Received 22 February 1996)

$$\int_0^\infty d\omega_a \, \frac{\Gamma_m(\mathbf{r},\omega_a) - \Gamma_0(\omega_a)}{\Gamma_0(\omega_a)} = 0$$

Integral and Local DOS

- For continuous dielectric media DOS is welldefined function of dimensionality and refraction index
- For certain mesoscopic structures like microcavities or photonic crystals DOS is understood
- In a general case of a complex inhomogeneous structures we have to introduce local DOS with a determination region on the order of lambda/n

Local DOS problem

"Begin by deciding how much of the universe needs to be brought into the discussion. Decide what normal modes are needed for an adequate treatment of the problem under consideration". Lamb W E Appl. Phys. B60 77 (1995)

PHYSICAL REVIEW E 69, 057601 (2004)

Electromagnetic density of modes for a finite-size three-dimensional structure

Giuseppe D'Aguanno,^{1,2,3,*} Nadia Mattiucci,^{1,2,4} Marco Centini,^{2,3} Michael Scalora,² and Mark J. Bloemer²

 "Quite surprisingly, the concept of the density of states for a finite-size structure still lacks a simple, concise definition"

$$\rho_{\omega} \equiv \rho_{\omega, \text{free space}} \frac{\langle W_{\text{emitted}}(\vec{r}) \rangle}{W_{\text{emitted}, \text{free space}}}$$
$$= -\frac{6\pi c}{\omega} \rho_{\omega, \text{free space}} \langle \text{Im}[G_{\omega, \hat{x}\hat{x}}(\vec{r}, \vec{r})] \rangle,$$

Sum rule for the local density of states

$$\int_{0}^{\infty} \frac{D(\mathbf{r}, \omega) - D_{free}(\omega)}{D_{free}(\omega)} d\omega = 0, \quad D_{free}(\omega) = \frac{\omega^{2}}{\pi^{2}c^{3}}$$

Clear analog in solid state physics: Impurity electron states in band gap develop by the expense of intrinsic electron states in c-band



Calculated photon density of states for a three-dimensional photonic crystals whose design and band structure were presented in Figure 7.23. Gray band indicates the band gap. Reprinted with permission from [70]. Copyright 2007, Elsevier B.V.

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PHYSICAL REVIEW E 77, 046602 (2008)

Constraints on transmission, dispersion, and density of states in dielectric multilayers and stepwise potential barriers with an arbitrary layer arrangement

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High transmission in complex structures correlates with higher DOS 54

Conclusion

- Since 1900 DOS contribution in quantum physics and optics has ben recognised
- Photon DOS contributes to emission and scattering of light providing strong enhancement in mesoscopic systems
- Total DOS can not be modified
- Local photon DOS needs further research