Multipole excitations in all-dielectric meta-molecules and in organic molecules

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Electric and magnetic fields described by the scalar and vector potentials

Electric and magnetic fields can be described by scalar and vector potentials:

$$\Phi(\boldsymbol{R},t)=\frac{1}{4\pi\varepsilon\varepsilon_0}\int_V \frac{\rho\left(\boldsymbol{r},t-\frac{|\boldsymbol{R}-\boldsymbol{r}|}{v}\right)}{|\boldsymbol{R}-\boldsymbol{r}|}dV$$

$$\boldsymbol{A}(\boldsymbol{R},t)=\frac{\mu\mu_0}{4\pi}\int_V \frac{\boldsymbol{J}\left(\boldsymbol{r},t-\frac{|\boldsymbol{R}-\boldsymbol{r}|}{v}\right)}{|\boldsymbol{R}-\boldsymbol{r}|}dV$$

Expansion of vector potential into Taylor series (scalar potential can be expanded with similar way):

$$\boldsymbol{A}(\boldsymbol{R},t)=\frac{\mu\mu_0}{4\pi}\int_V \frac{\boldsymbol{J}\left(\boldsymbol{r},t-\frac{|\boldsymbol{R}-\boldsymbol{r}|}{v}\right)}{|\boldsymbol{R}-\boldsymbol{r}|}dV=$$

$$=\frac{\mu\mu_0}{4\pi R}\left[\int_V \boldsymbol{J}\,dV+\frac{R_i}{vR}\int_V \dot{\boldsymbol{J}}r_i\,dV+\frac{R_jR_k}{2v^2R^2}\int_V \ddot{\boldsymbol{J}}r_jr_k\,dV+\frac{R_jR_kR_m}{6v^3R^3}\int_V \dddot{\boldsymbol{J}}r_jr_kr_m\,dV+\ldots\right]$$

Development of amendments to classical multipole decomposition

Consideration of the second term in vector potential leads us to the term of the form:

$$R_j \int_V J_i r_j dV$$

To treat it we will use the following auxiliary expression:

$$R_j \int_V \nabla \left(\boldsymbol{J} r_i r_j \right) dV = R_j \int_V \left(\dot{\rho}\, r_i r_j + J_i R_j r_j + r_i R_j J_j \right) dV =$$

$$= -\dot{Q}_{ij} R_j + 2 \int_V J_i \left(R_j r_j \right) dV + \int_V \underbrace{\left[r_i (R_j J_j) - J_i (R_j r_j) \right]}_{\boldsymbol{b}(\boldsymbol{ac}) - \boldsymbol{c}(\boldsymbol{ab}) = [\boldsymbol{a} \times [\boldsymbol{b} \times \boldsymbol{c}]]} dV =$$

$$= -\dot{Q}_{ij} R_j + 2 \int_V J_i \left(R_j r_j \right) dV + \boldsymbol{R} \times \int_V \left[\boldsymbol{r} \times \boldsymbol{J} \right] dV$$

And finally obtain:

$$R_j \int_V J_i r_j dV = \frac{1}{2} \dot{Q}_{ij} R_j + \left[\boldsymbol{m} \times \boldsymbol{R} \right] + \frac{1}{2} U'_{ij} R_j$$

Multipole moments

Electric multipole moments:

$$d_i = \int_V \rho(\boldsymbol{r}) r_i \, dV = \frac{1}{i\omega}\left(U_i + \int_V J_i \, dV\right)$$

$$Q_{ij} = \int_V \rho(\boldsymbol{r})\, r_i r_j \, dV = \frac{1}{i\omega}\left(U' + \int_V (J_i r_j + J_j r_i)\, dV\right)$$

$$O_{ijk} = \int_V \rho(\boldsymbol{r})\, r_i r_j r_k \, dV = \frac{1}{i\omega}\left(U'' + \int_V (J_i r_j r_k + r_i J_j r_k + r_i r_j J_k)\, dV\right)$$

Magnetic multipole moments:

$$\boldsymbol{m} = \frac{1}{2}\int_V [\boldsymbol{r} \times \boldsymbol{J}]\, dV$$

$$M_{qm} = \frac{2}{3}\int_V [\boldsymbol{r} \times \boldsymbol{J}]_q \, r_m \, dV$$

Surface integrals:

$$U_i = \oint_S (\boldsymbol{n}_S \cdot \boldsymbol{J})\, r_i \, dS$$

$$U'_{ij} = \oint_S (\boldsymbol{n}_S \cdot \boldsymbol{J})\, r_i r_j dS$$

$$U''_{ijk} = \oint_S (\boldsymbol{n}_S \cdot \boldsymbol{J})\, r_i r_j r_k \, dS$$

Multipole decomposition technique

Multipole decomposition of vector potential:

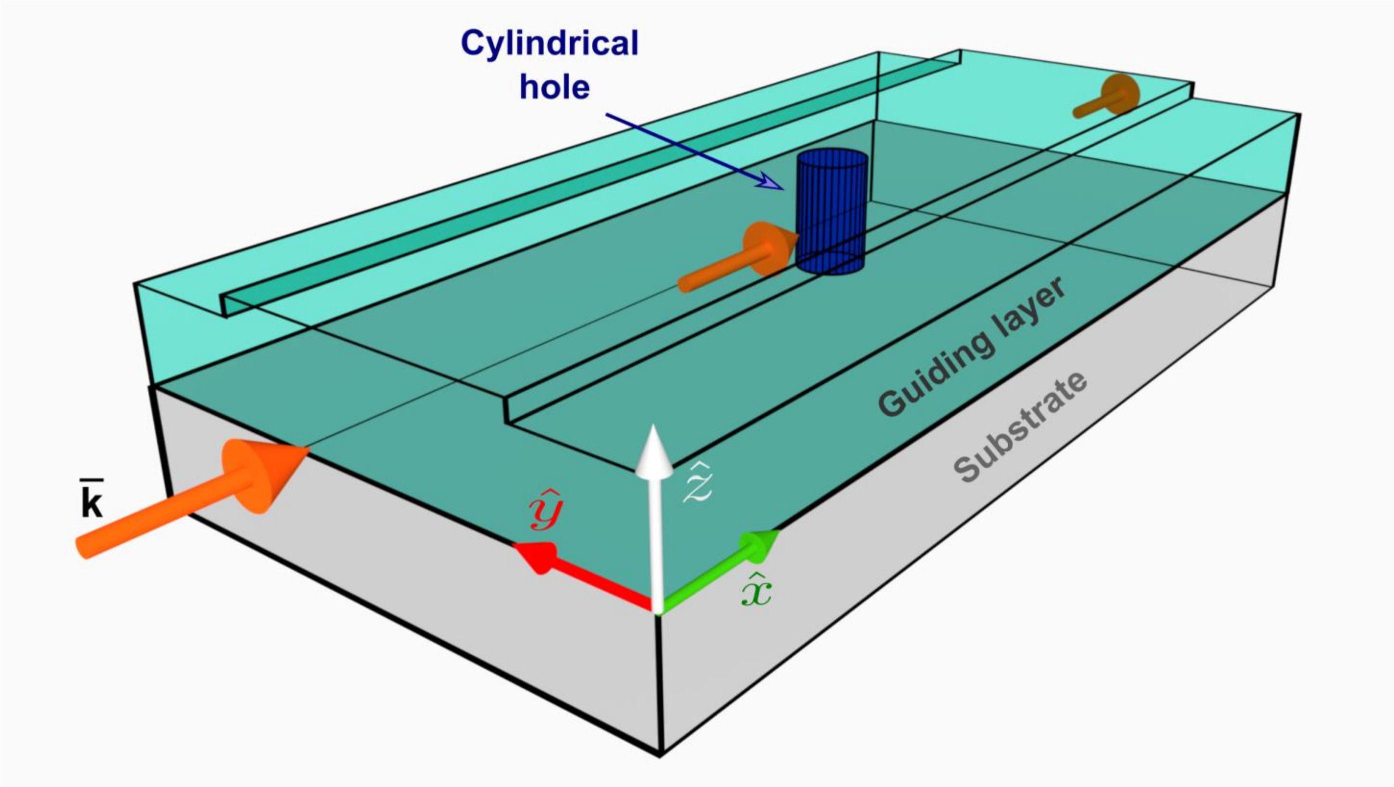
$$\boldsymbol{A}(\boldsymbol{R},t)=\frac{\mu\mu_0}{4\pi R}\left[\dot{\boldsymbol{d}}+\boldsymbol{U}+\frac{1}{2v}\ddot{Q}\boldsymbol{n}+\frac{1}{v}\left[\dot{\boldsymbol{m}}\times\boldsymbol{n}\right]+\frac{1}{2v}\dot{U}'\boldsymbol{n}+\frac{1}{6v^2}\dddot{O}\boldsymbol{n}\boldsymbol{n}+\right.$$
$$\left.+\frac{1}{2v^2}\left[\boldsymbol{n}\times\ddot{M}\boldsymbol{n}\right]+\frac{1}{6v^2}\ddot{U}''\boldsymbol{n}\boldsymbol{n}+\dots\right]$$

Multipole decomposition of magnetic and electric fields:

$$\boldsymbol{H}=\frac{\mu\mu_0}{4\pi R v}\frac{e^{ikR}}{R}\left(\left[\ddot{\boldsymbol{d}}\times\boldsymbol{n}\right]+\left[\dot{\boldsymbol{U}}\times\boldsymbol{n}\right]+\frac{1}{2v}\left[\dddot{Q}\boldsymbol{n}\times\boldsymbol{n}\right]+\frac{1}{v}\left[\boldsymbol{n}\times\left[\boldsymbol{n}\times\ddot{\boldsymbol{m}}\right]\right]+\right.$$
$$\left.+\frac{1}{2v}\left[\ddot{U}'\boldsymbol{n}\times\boldsymbol{n}\right]+\frac{1}{6v^2}\left[\ddddot{O}\boldsymbol{n}\boldsymbol{n}\times\boldsymbol{n}\right]+\frac{1}{2v^2}\left[\boldsymbol{n}\times\left[\dddot{M}\boldsymbol{n}\times\boldsymbol{n}\right]\right]+\frac{1}{6v^2}\left[\dddot{U}''\boldsymbol{n}\boldsymbol{n}\times\boldsymbol{n}\right]\right)$$

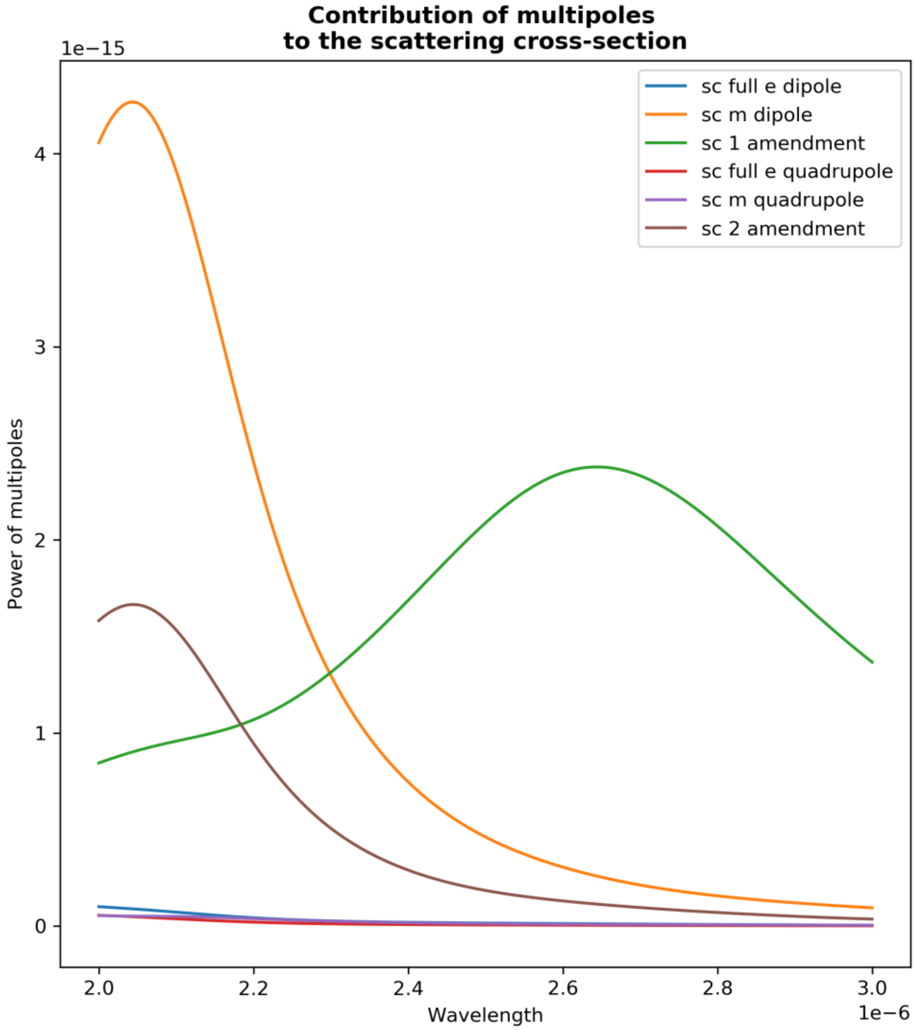
$$\boldsymbol{E}=\frac{k^2}{4\pi\varepsilon\varepsilon_0}\frac{e^{ikR}}{R}\left(\left[\boldsymbol{n}\times\left[\boldsymbol{d}\times\boldsymbol{n}\right]\right]+\frac{i}{kv}\left[\boldsymbol{n}\times\left[\boldsymbol{U}\times\boldsymbol{n}\right]\right]+\frac{ik}{2}\left[\boldsymbol{n}\times\left[Q\boldsymbol{n}\times\boldsymbol{n}\right]\right]+\frac{1}{v}\left[\boldsymbol{m}\times\boldsymbol{n}\right]+\right.$$
$$\left.+\frac{1}{2v}\left[\boldsymbol{n}\times\left[U'\boldsymbol{n}\times\boldsymbol{n}\right]\right]+\frac{k^2}{6}\left[\boldsymbol{n}\times\left[\boldsymbol{n}\times O\boldsymbol{n}\boldsymbol{n}\right]\right]+\frac{ik}{2v}\left[\boldsymbol{n}\times M\boldsymbol{n}\right]+\frac{ik}{6v}\left[\boldsymbol{n}\times\left[\boldsymbol{n}\times U''\boldsymbol{n}\boldsymbol{n}\right]\right]\right)$$

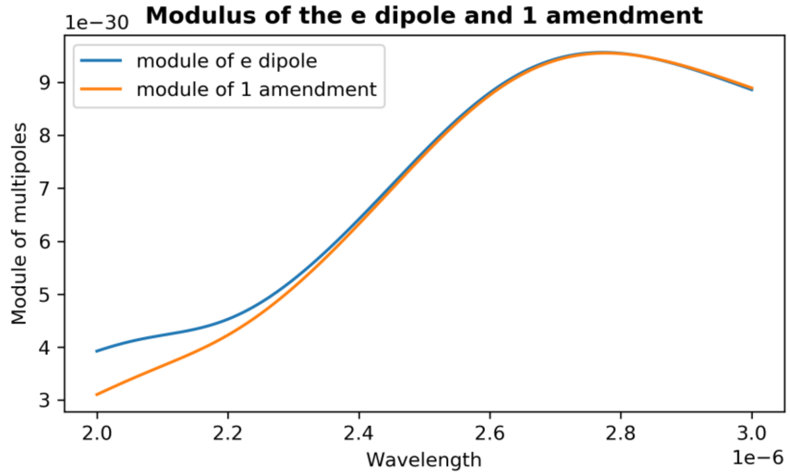
The open system with meta-molecule: optical waveguide with inclusion index of n=1

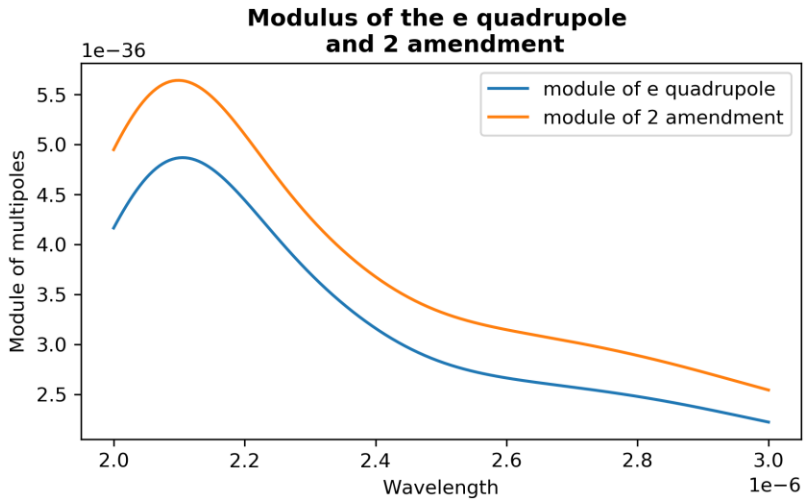


Silicon on insulator waveguide with inclusion of cylindrical shape and index of n=1. In this case the integrated surface cannot be placed at infinity, where electric and magnetic fields are zero.

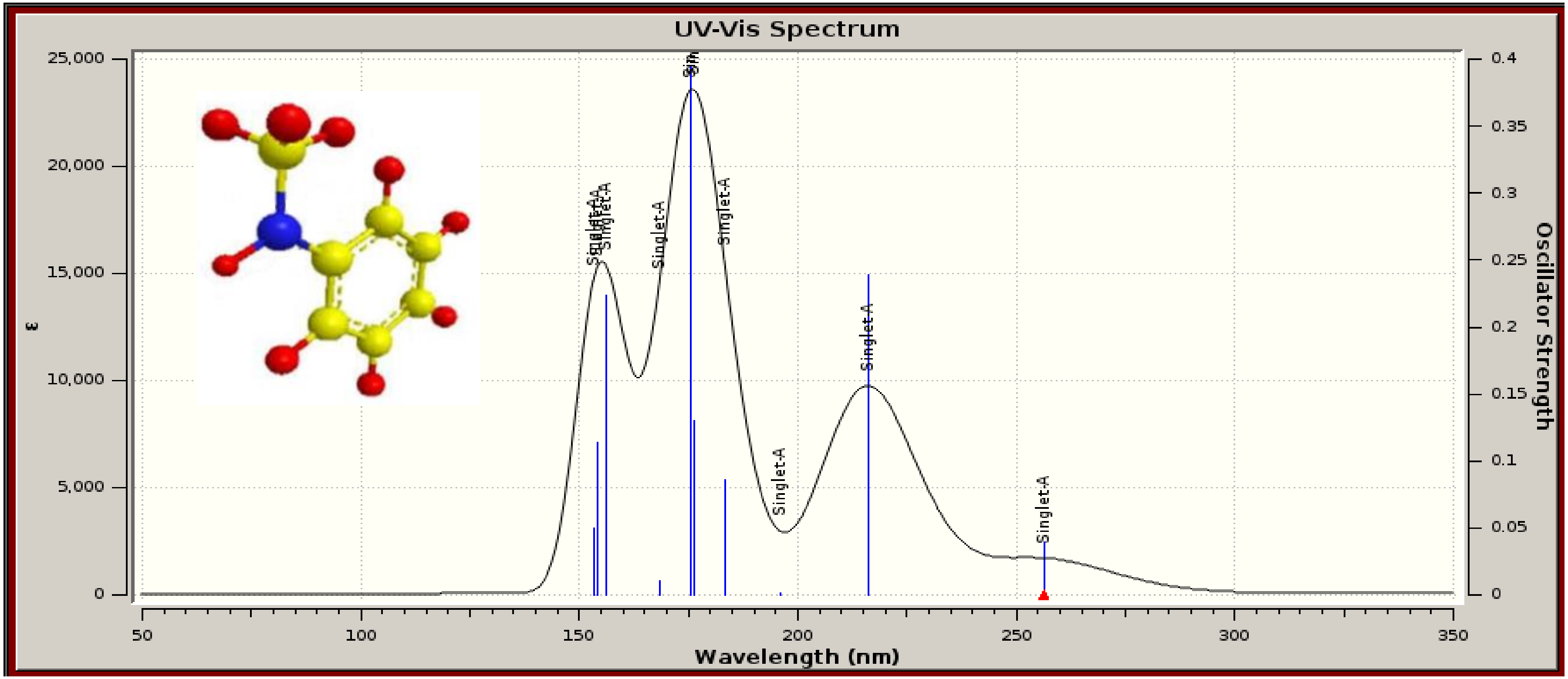
Multipole decomposition of considered inclusion







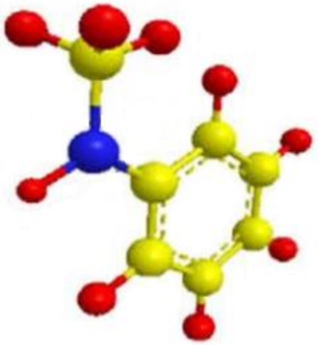
Multipoles of organic molecule: N-Methylaniline



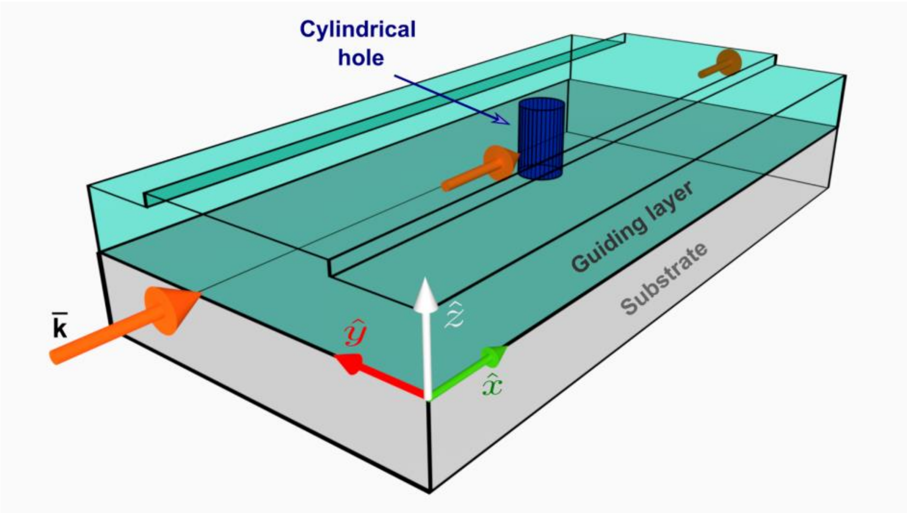
Ground state: d = 2,0512
1 exited state: d = 1,7813
2 exited state: d = 1,7812
3 exited state: d = 2,0106
4 exited state: d = 0,2864

5 exited state: d = 0,6706
6 exited state: d = 0,286
7 exited state: d = 1,5433
8 exited state: d = 0,9966
9 exited state: d = 0,671

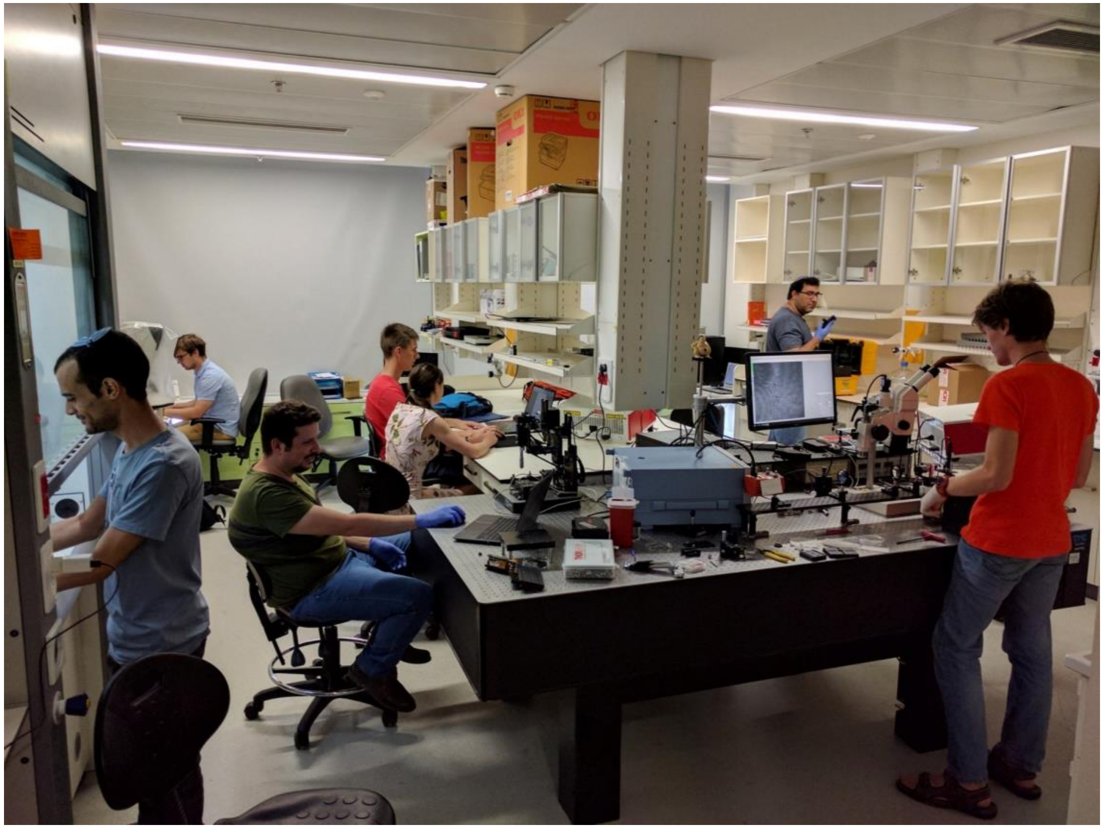
Conclusion



- **Amendments** were proposed to the classical multipole decomposition to allow for analysis of open systems in which charges and currents are not localised.
- In case of an **inclusion filled with** air on an optical waveguide, electric multipole moments are replaced by the surface integrals.
- **Molecular multipoles** can be analysed by the classical multipole decomposition. Only electric molecular multipoles can exist in most of the conventional molecules to the best of our knowledge.



Thank you for your attention!



Our team in Ben-Gurion University