Normalizing $\chi(-2\omega;\omega,\omega)$ for Specific Cases

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First Case: Bulk Materials

Let us first consider the case of a bulk material, that is conformed entirely of a supercell with no vacuum region. A unit cell is repeated indefinitely in every direction, thus creating a three-dimensional reproduction of an infinite material. From Fig. 1, we can see that the volume of each unit cell is $\Omega = L^3$. We calculate $\chi(-2\omega;\omega,\omega)$ using the TINIBA [1] software suite; for ease of notation, $\chi(-2\omega;\omega,\omega) = \chi_T$ (T for TINIBA).

For the bulk calculation, χ_T must be normalized over the volume of the unit cell (i.e. $1/\Omega$); this normalization happens automatically in TINIBA. Thus, no further action is necessary and the susceptibility can be used as is. Finally, we establish that $\chi_T = \chi_b$, where χ_b is the desired susceptibility for our bulk system. Calculated in this fashion, χ_b has the appropriate MKS units of m/V.

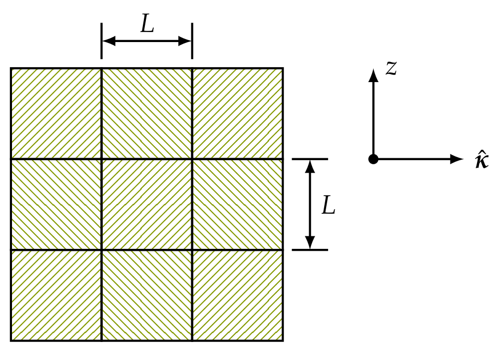
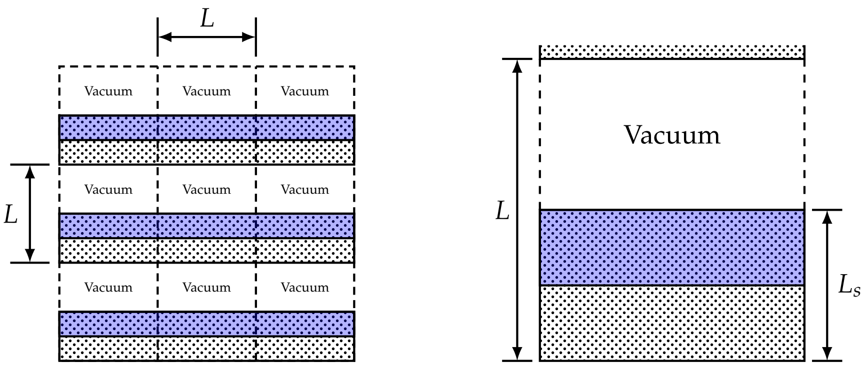


Figure 1: The bulk supercell is repeated indefinitely along the z and $\hat{\kappa}$ axes. There is only material present, with no vacuum region between each repeat.

Second Case: Surfaces

Now, we will consider calculating $\chi(-2\omega;\omega,\omega)$ for surfaces. This is done following the theoretical framework established in Ref. [2]. Just as for the bulk case, we use the supercell method that repeats each cell across all directions (see Fig. 2). However, we represent the surface by using a slab of material with finite height; this necessarily implies that there are regions of empty space between each repeat. The slab has both upper and lower surfaces, and we can extract the response for each by use of the cut function.



(a) Repeated supercells for a surface material. Each individual supercell is repeated in every direction, just like the bulk case. Vacuum regions must be included, seperating each supercell.

(b) An individual supercell, which consists of a slab (with upper and bottom surfaces) and a vacuum region. The total height will be the combined height of the vacuum region and the slab (L_s).

Figure 2: Just like for the bulk case, we use the supercell scheme for calculating $\chi(-2\omega;\omega,\omega)$ for surfaces.

Fig. 2b depicts a representative supercell for a surface material. As mentioned above, it is necessary to include the cut-function to extract the surface response. For instance, a centrosymmetric material will always yield $\chi(-2\omega;\omega,\omega) = 0$, except at the surface where the symmetry is broken. If we calculate the response from the entire slab, the contribution from the bottom half will cancel out the top half; thus, we use the cut function to only calculate over the desired region. For most surfaces, we can simply

The blue shaded region represents the *half-slab*

Third Case: Two-dimensional Materials

References

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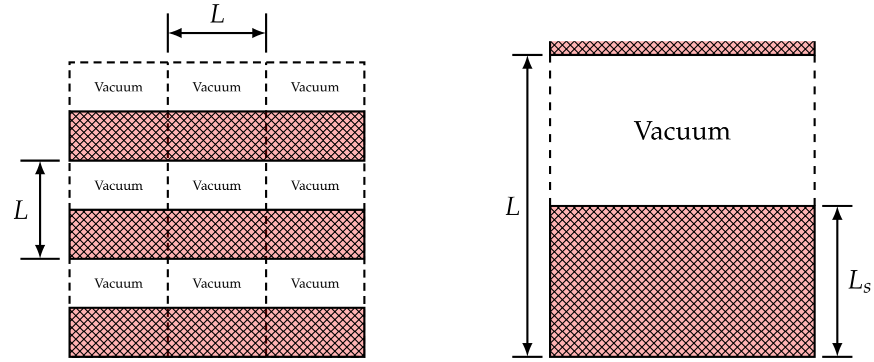

(a) Repeated supercells for a 2D material. Again, each individual supercell is repeated in every direction. Vacuum regions seperate each slab of material.

(b) An individual supercell, with slab and vacuum region. We calculate the response from the entire slab, disregarding any layers.

Figure 3: The supercell scheme for calculating $\chi(-2\omega;\omega,\omega)$ for 2D materials is very similar to the surface case.

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[2] S. M. Anderson, N. Tancogne-Dejean, B. S. Mendoza, and V. Véniard. Theory of surface second-harmonic generation for semiconductors including effects of nonlocal operators. *Physical Review B*, 91(7):075302, February 2015.