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Symmetry Assumptions, Kramers–Kronig Transformation and Analytical Continuation in *Ab Initio* Calculations of Optical Conductivities

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Abstract

The limit of infinite relaxation of the Kubo formula and analytical and numerical properties of the Kramers–Kronig transformation and analytical continuation used in *ab initio* calculations of the optical conductivity tensor are considered. Essential symmetry assumptions used in magneto optics are pointed out and their validity for some classes of important systems is shown. It is shown that for an energy dependent relaxation time, the optical conductivity can always be calculated with desired numerical accuracy by applying a Kramers–Kronig transformation and analytical continuation to the result obtained in the limit of infinite relaxation time instead of calculating it directly from the Kubo formula with a finite relaxation time. Consequently the difference between the two approaches is reduced to the difference between the Brillouin zone integration techniques.

1. Introduction

Advanced by the interest in magneto optical data storage, the calculation of magneto optical effects by means of band structure methods has become a growing branch in the field of *ab initio* computational physics. The commonly used computational sceme [1–4] was proposed by Wang and Callaway [5,6] in 1974. It is based on the calculation of the dielectric function or equivalently the optical conductivity tensor in linear response theory from the Kubo–Greenwood [7] formalism. However, considering magneto optics, most implementations in currently used band structure programs such as, e.g., the WIEN2k [8] code stress symmetry assumptions that have not been fully discussed in the literature. Further, they demand additional operations, namely Kramers–Kronig transformation and analytical continuation whose validity and numerical accuracy has been the issue of open discussions [9].

In this article we discuss the role of the symmetry assumptions used in magneto optics and recall that they are valid for many systems of physical interest. We point out that the Kramers–Kronig transformation and analytical continuation can always be carried out with desired accuracy and give expressions that can be implemented numerically. The article is organized as follows: In Section 2 we review the derivation of the limit $\tau \longrightarrow \infty$ of the Kubo formula and show how symmetry assumptions are used to obtain the commonly used simplified expression. In Section 3 we show how both Kramers–Kronig transformation and analytical continuation can be calculated numerically with arbitary accuracy. In Section 4 we consider some classes of physical systems that satisfy the acquired assumptions. Finally in Section 5 we summarize our considerations.

2. Infinite lifetime limit of the Kubo formula

If we consider only direct inter band transitions, the optical conductivity tensor in linear response theory is obtained from the Kubo–Greenwood formalism by [5,10]

$$\sigma_{\alpha\beta}(\omega) = \frac{\mathrm{i}e^2}{m^2\hbar}\int_{BZ} \mathrm{d}^3k \sum_{\substack{l,n \\ E_l(\boldsymbol{k})<E_F \\ E_n(\boldsymbol{k})<E_F}} \frac{1}{\omega_{nl}(\boldsymbol{k})} \times \left[\frac{\Pi^{\alpha}_{ln}(\boldsymbol{k})\Pi^{\beta}_{nl}(\boldsymbol{k})}{\omega-\omega_{nl}(\boldsymbol{k})+\frac{i}{\tau(\omega)}} + \frac{(\Pi^{\alpha}_{ln}(\boldsymbol{k})\Pi^{\beta}_{nl}(\boldsymbol{k}))^*}{\omega+\omega_{nl}(\boldsymbol{k})+\frac{\mathrm{i}}{\tau(\omega)}}\right]. \quad (1)$$

The indices l and n denote the spin and all band quantum numbers for the occupied and empty states respectively and $\boldsymbol{k}$ is the continuous quantum number related to the translational symmetry and restricted to the Brillouin zone, E_F is the Fermi energy. The symbol $\Pi^{\alpha}_{nl}(\boldsymbol{k})$, $\alpha = x, y, z$ denotes the matrix elements of the momentum operator given below by Eq. (3) and $\omega_{nl}(\boldsymbol{k})$ is the energy difference between the involved states.

$$\omega_{nl}(\boldsymbol{k}) = \frac{1}{\hbar}(E_n(\boldsymbol{k}) - E_l(\boldsymbol{k})). \quad (2)$$

Finally $\tau(\omega)$ is a phenomenological relaxation time.

The matrix elements of the momentum operator are obtained from a band structure calculation by evaluating the expression

$$\boldsymbol{\Pi}_{ln}(\boldsymbol{k}) = \int \mathrm{d}^3r\ \psi^*_l(\boldsymbol{k},\boldsymbol{r})\left[\boldsymbol{p} + \frac{\hbar}{4mc^2}[\boldsymbol{\sigma}\times\boldsymbol{\nabla}V(\boldsymbol{r})]\right] \times \psi_n(\boldsymbol{k},\boldsymbol{r}). \quad (3)$$

Here $\psi_n(\boldsymbol{k},\boldsymbol{r})$ is the Bloch wave function with quantum numbers as described above, $\boldsymbol{p} = -\mathrm{i}\hbar\boldsymbol{\nabla}$, $V(\boldsymbol{r})$ is a crystal potential. Note that the second term in the square brackets describing the contribution of the spin–orbit coupling to the matrix elements is usually taken into account by approximative scalar relativistic methods such as the second variational step or first order pertubation theory [5]. However, when magneto optical effects are considered, in principle the full expression should be calculated.

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We take the limit of infinite relaxation time in the integral over the Brillouin zone that appears in Eq. (1), i.e, for any ω, we let $\tau(\omega)\longrightarrow\infty$. Using [11]

$$\lim_{\varepsilon\to 0}\int \mathrm{d}x \frac{1}{x+\mathrm{i}\varepsilon} f(x) = -\mathrm{i}\pi\int \mathrm{d}x\ \delta(x)f(x) + \mathcal{P}\!\!\!\!\!\!\int \mathrm{d}x \frac{f(x)}{x} \tag{4}$$

where the P denotes the principle value integral, we get

$$\begin{aligned}\sigma_{\alpha\beta}(\omega) = \frac{\mathrm{i}e^2}{m^2\hbar}\Bigg[&-\mathrm{i}\pi\int_{BZ}\mathrm{d}^3k \sum_{\substack{l,n\\ E_l(\boldsymbol{k})<E_F\\ E_n(\boldsymbol{k})<E_F}} \frac{1}{\omega_{nl}(\boldsymbol{k})}\\ &\times\Big[\Pi^{\alpha}_{ln}(\boldsymbol{k})\Pi^{\beta}_{nl}(\boldsymbol{k})\delta(\omega-\omega_{nl}(\boldsymbol{k}))\\ &+\Big(\Pi^{\alpha}_{ln}(\boldsymbol{k})\Pi^{\beta}_{nl}(\boldsymbol{k})\Big)^{*}\delta(\omega+\omega_{nl}(\boldsymbol{k}))\Big]\\ &+\mathcal{P}\!\!\!\!\!\!\int_{BZ}\mathrm{d}^3k \sum_{\substack{l,n\\ E_l(\boldsymbol{k})<E_F\\ E_n(\boldsymbol{k})<E_F}} \frac{1}{\omega_{nl}(\boldsymbol{k})}\\ &\times\Bigg[\Pi^{\alpha}_{ln}(\boldsymbol{k})\Pi^{\beta}_{nl}(\boldsymbol{k})\frac{1}{\omega-\omega_{nl}(\boldsymbol{k})}\\ &+\Big(\Pi^{\alpha}_{ln}(\boldsymbol{k})\Pi^{\beta}_{nl}(\boldsymbol{k})\Big)^{*}\frac{1}{\omega+\omega_{nl}(\boldsymbol{k})}\Bigg]\Bigg].\end{aligned} \tag{5}$$

The last term in the first line of Eq. (5) immediately drops out, if we are only interested in the optical conductivity in the positive energy region. Since the operator (3) is hermitian, one gets

$$(\Pi^{\alpha}_{ln}\Pi^{\beta}_{nl})^{*} = \Pi^{\beta}_{ln}\Pi^{\alpha}_{nl} \tag{6}$$

thus for $\beta=\alpha$ one has $\mathrm{Im}[\Pi^{\alpha}_{ln}\Pi^{\alpha}_{nl}]=0$ and the real and imaginary parts of Eq. (5) contain the integrals over the δ-function and the principle value integrals respectively. If we consider only the real part of the conductivity, the principle value integrals drop out and we obtain the expression that is most commonly implemented in bandstructure calculations [10].

$$\begin{aligned}\mathrm{Re}[\sigma_{\alpha\alpha}(\omega)] = \frac{e^2\pi}{m^2\hbar\omega}\int_{BZ}\mathrm{d}^3k.\\ \times\sum_{\substack{l,n\\ E_l(\boldsymbol{k})<E_F\\ E_n(\boldsymbol{k})<E_F}} |\Pi^{\alpha}_{ln}(\boldsymbol{k})|^2\delta(\omega-\omega_{nl}(\boldsymbol{k})).\end{aligned} \tag{7}$$

Analoguous arguments do not in general hold for the off diagonal components of the optical conductivity tensor, as both real and imaginary part contain principle value integrals. However it will be demonstrated in Section 4 that the off diagonal elements of the conductivity tensor are antisymmetric for many systems of interest in magneto optics.

Stressing this property we can replace in Eq. (5)

$$\begin{aligned}\Pi^{\alpha}_{ln}(\boldsymbol{k})\Pi^{\beta}_{nl}(\boldsymbol{k})\longmapsto&\frac{1}{2}\Big[\Pi^{\alpha}_{ln}(\boldsymbol{k})\Pi^{\beta}_{nl}(\boldsymbol{k})-\Pi^{\beta}_{ln}(\boldsymbol{k})\Pi^{\alpha}_{nl}(\boldsymbol{k})\Big]\\ &\equiv \mathrm{i}\,\mathrm{Im}[\Pi^{\alpha}_{ln}(\boldsymbol{k})\Pi^{\beta}_{nl}(\boldsymbol{k})]\end{aligned} \tag{8}$$

where we have taken into account Eq. (6). We obtain the formula for the off diagonal components of the optical conductivity tensor that is commonly used in magneto optics.

$$\begin{aligned}\mathrm{Im}[\sigma_{\alpha\beta}(\omega)] = \frac{\pi e^2}{m^2\hbar\omega}\int_{BZ}\mathrm{d}^3k\\ \times\sum_{\substack{l,n\\ E_l(\boldsymbol{k})<E_F\\ E_n(\boldsymbol{k})<E_F}}\mathrm{Im}[\Pi^{\alpha}_{ln}\Pi^{\beta}_{nl}]\delta(\omega-\omega_{nl}(\boldsymbol{k})).\end{aligned} \tag{9}$$

Equations (7) and (9) can be implemented numerically much more efficiently than Eq. (1) because only the fast converging contributions of the Brillouin zone integrations have to be calculated [12]. Yet, both for the diagonal and for the off diagonal components of the optical conductivity tensor, only one part of the full complex expression is obtained. Thus a successive Kramers–Kronig transformation is necessary to obtain the imaginary and real parts respectively. As we are interested in the optical conductivity tensor for a finite lifetime $\tau(\omega)$, we also have to make an analytical continuation the result we obtained from the limit of infinite relaxation time.

3. Kramers–Kronig transformation and analytical continuation

The real and imaginary parts of the optical conductivity in linear response theory are related by Kramers–Kronig transformations

$$\sigma_1(\omega) = \frac{2}{\pi}\mathcal{P}\!\!\!\!\!\!\int_0^{\infty}\frac{\omega'\sigma_2(\omega')}{\omega'^2-\omega^2}\,\mathrm{d}\omega', \tag{10}$$

$$\sigma_2(\omega) = -\frac{2\omega}{\pi}\mathcal{P}\!\!\!\!\!\!\int_0^{\infty}\frac{\sigma_1(\omega')}{\omega'^2-\omega^2}\,\mathrm{d}\omega' \tag{11}$$

(see, e.g., Ref. [13]). They are valid for any complex function defined in the complex plane satisfies the condition

$$\sigma(-z^*) = \sigma^*(z) \tag{12}$$

has no pole in the upper complex half plane and is decaying sufficiently fast to zero if ω goes to infinity on any path in the upper complex half plane.

Kramers–Kronig relations are valid for an exact conductivity tensor. Now we should prove that standard approximations made in the band structure calculation do not violate them. The condition Eq. (12) is satisfied by both Eqs. (1) and (5) and the calculated optical conductivities are obviously analytical in the upper complex halfplane. It remains to show that they are decaying to zero in the upper complex half plane and a proper treatment of the infinite integrals needs to be found. In any band structure calculation only a finite number of basis functions is used and there will be some maximum energy difference $\omega_{nl}(\boldsymbol{k})$ that basically corresponds to the difference in energy between the lowest and the highest states that are covered by the basis set. Likewise, there will be a finite set of matrix elements of the momentum operator which contributes only in the energy range that is covered by the basis set and vanishes very rapidly outside. Thus the optical conductivity tensor calculated from Eq. (1) or Eq. (5) goes to zero for $\omega\longrightarrow\infty$ and is in fact numerically identical to zero above some finite energy $\omega_{\max}$ that we can

roughly estimate from the basis set used in the band structure calculation. Note that this is essentially different from any real physical optical conductivity which has resonant peaks up to very high energies.

Suppose an optical conductivity $\sigma(\omega)$ satisfies Eq. (12) and is identically zero for $|\omega| > \omega_{\max}$. Using

$$\mathcal{P}\int_0^\infty \frac{1}{\omega'^2 - \omega^2}\,\mathrm{d}\omega' = 0 \tag{13}$$

and

$$\mathcal{P}\int_{\omega_{\max}}^\infty \frac{1}{\omega'^2 - \omega^2}\,\mathrm{d}\omega' = \frac{1}{2}\ln\left|\frac{\omega+\omega_{\max}}{\omega-\omega_{\max}}\right|, \qquad \omega < \omega_{\max} \tag{14}$$

one obtains for the Kramers–Kronig relations

$$\sigma_1(\omega) = \frac{2}{\pi}\mathcal{P}\int_0^{\omega_{\max}} \frac{\omega'\sigma_2(\omega') - \omega\sigma_2(\omega)}{\omega'^2 - \omega^2}\,\mathrm{d}\omega' - \omega\sigma_2(\omega) \times \frac{1}{\pi}\ln\left|\frac{\omega+\omega_{\max}}{\omega-\omega_{\max}}\right| \tag{15}$$

and

$$\sigma_2(\omega) = -\frac{2\omega}{\pi}\mathcal{P}\int_0^{\omega_{\max}} \frac{\sigma_1(\omega') - \sigma_1(\omega)}{\omega'^2 - \omega^2}\,\mathrm{d}\omega' - \omega\sigma_1(\omega) \times \frac{1}{\pi}\ln\left|\frac{\omega+\omega_{\max}}{\omega-\omega_{\max}}\right| \tag{16}$$

for $\omega < \omega_{\max}$. Replacing the integrands by their limits at the undefined points, the evaluation of the principle values is obsolete and the integrations can be performed numerically with standard interpolation techniques. Note that the expression is undefined for $\omega = \omega_{\max}$ and numerically less accurate in a small region close to this point. Yet, in the region we are interested in, where $\sigma_1(\omega)$ and $\sigma_2(\omega)$ are nonvanishing, the accuracy depends only on the mesh density and interpolation technique used in the integration routine. Hence, choosing a sufficiently big $\omega_{\max}$ and an appropriate mesh, we can calculate Kramers–Kronig transformations with any desired accuracy for any optical conductivity obtained with Eq. (1) or Eq. (5) from a band structure calculation.

Suppose $\sigma^\infty(\omega)$ is an optical conductivity in the limit of infinite relaxation calculated from Eq. (5) or the simplified expressions Eq. (7) and (9). Further let $\tau(\omega)$ be the phenomenological relaxation time of the system. Then the optical conductivity $\sigma^{\tau(\omega)}(\omega)$ with the real relaxation time given by the Kubo formula Eq. (1) is related to $\sigma^\infty(\omega)$ by

$$\sigma^{\tau(\omega)}(\omega) = \sigma^\infty\left(\omega + \frac{\mathrm{i}}{\tau(\omega)}\right) \tag{17}$$

i.e., by an analytical continuation $\omega \longmapsto \omega + \mathrm{i}/\tau(\omega)$.

Equation (17) coincides with the result of a convolution of the optical conductivity with a normalized Lorentzian of width $\eta(\omega) = 1/\tau(\omega)$,

$$\int_{-\infty}^\infty \sigma^\infty(\omega')\frac{1}{\pi}\frac{\eta(\omega)}{(\omega-\omega')^2 + \eta(\omega)^2}\,\mathrm{d}\omega'. \tag{18}$$

As $\sigma(\omega) \longrightarrow 0$ for $\omega \longrightarrow \infty$ on any path in the upper complex half plane, we may rewrite this as a complex integral over the real axis and an infinite half circle

$$= \lim_{R\to\infty} \oint \sigma^\infty(z)\frac{1}{\pi}\frac{\eta(\omega)}{(\omega-z)^2 + \eta(\omega)^2}\,\mathrm{d}z \tag{19}$$

which can be written as a sum over the residuals in the enclosed area, that is the upper half plane. Recalling that $\sigma^\infty(z)$ does not have any poles in the upper complex half plane we find only one residual at $z = \omega + \mathrm{i}\eta(\omega)$. As a result we get

$$\int_{-\infty}^\infty \sigma^\infty(\omega')\frac{1}{\pi}\frac{\eta(\omega)}{(\omega-\omega')^2 + \eta(\omega)^2}\,\mathrm{d}\omega' = \sigma^\infty(\omega + \mathrm{i}\eta(\omega)). \tag{20}$$

The integrand is everywhere well defined and as argued above, it vanishes for $|\omega| > \omega_{\max}$. By Eq. (12) we obtain the integrand in the negative energy range. Thus Eq. (20) can be implemented numerically. Having choosen an appropriate $\omega_{\max}$, the accuracy depends once again only on the mesh density and interpolation technique used in the integration routine.

Note that the analytical continuation obtained with Eq. (20) can be calculated for the real and imaginary part separately, i.e., we might apply it only to the real or imaginary part of an optical conductivity calculated by Eq. (7) or Eq. (9). The result we get corresponds to the real or imaginary part of the optical conductivity we would have obtained from Eq. (1). As Eq. (1) satisfies the conditions for Kramers–Kronig transformation, the analytically continued real and imaginary parts must satisfy them as well. Thus we can apply Kramers–Kronig transformation also to the analytically continued real or imaginary part of the optical conductivity and we obtain the full complex conductivity given by Eq. (1). That means that it does not matter if we first perform a Kramers–Kronig transformation and subsequently calculate the analytical continuation or the other way round. This suggests to use the comparison between the results obtained in both ways as a check for the parameters used in the numerical implementations. In particular the cut off energy $\omega_{\max}$, which is usually difficult to estimate, can be verified in this way. It was found earlier [9] in numerical tests by A. Delin that Kramers–Kronig transformation and analytical continuation did not commute. However, in that work the author considered the relaxation time to be a function of the integration variable rather than a function of the energy for which the analytical continuation is calculated. Further, a sum of two antisymmetrically centered δ-distributions was used as a model conductivity. This is not an analytic function in the upper complex halfplane. The "analytical continuation" of the δ-distribution into the upper complex plane was calculated by a convolution with a lorentzian rather than by taking it to be identically zero. It remains an open question how the results of that work are to be understood.

4. Symmetry of the optical conductivity tensor

The approach to calculate conductivity tensors for magneto optics from band structure methods discussed in this paper is, as we pointed out in Section 2, based on the assumption $\sigma_{\alpha\beta} = -\sigma_{\beta\alpha}$ for $\alpha \neq \beta$. For a given crystal

structure and magnetisation direction, the symmetry properties of the conductivity tensor should in principle be determinable by identifying the crystal with one of the 90 magnetic symmetry classes [14,15] and examining the invariance of the wave functions and momentum operator under the symmetry operations. As rotations of the spin and spin–orbit coupling may be involved, this appears to be a non-trivial task in the general case. However, if the point group contains rotations around the spin axis, the above symmetry assumption may be verified by very basic considerations.

If the crystal has threefold or fourfold rotation symmetry around the magnetization direction [16] and the magnetization direction is taken as the z-axis, we obtain

$$\sigma = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} & 0 \\ -\sigma_{xy} & \sigma_{xx} & 0 \\ 0 & 0 & \sigma_{zz} \end{pmatrix}. \tag{21}$$

Thus the above symmetry assumption is satisfied. This is the well known symmetry appearing in basically all calculations of conductivity tensors for magneto optics from *ab initio* methods and it covers most cases of physical interest. For example, cubic Fe, Co and Ni with the magnetization along the main crystal axis as well as the cube diagonal or tetragonal and hcp atomic crystals with the magnetization along the z-axis.

5. Summary and conclusions

We have considered the most common approach used to calculate conductivity tensor for magneto optics from *ab initio* methods. It is based on the limit of infinite relaxation time of the Kubo formula Eq. (1), Kramers–Kronig transformation and analytical continuation via convolution with a Lorentzian. We acquired the symmetry assumptions about the optical conductivity that are necessary to perform this approach and pointed out some systems for which they are fulfilled. Further we showed that the assumptions needed to apply Kramers–Kronig transformation and analytical continuation are fulfilled by any *ab initio* calculated optical conductivity. For the related integrals, we presented modified expressions that can be implemented numerically and may be evaluated with any desired accuracy.

It may be concluded that the discussed approach is accurate for systems satisfying the required symmetry assumptions. Comparing the approach to the direct calculation of the optical conductivity from Eq. (1), we note that it gives the same result provided that the Brillouin zone integrations appearing in the respective Kubo formulas Eq. (1) and (7) or Eq. (9) are evaluated accurately. Thus the substantial difference between both methods reduces to the difference in the Brillouin zone integration techniques. As has been pointed out before [10], their accuracy depends strongly on the number of $\boldsymbol{k}$-points used in the band structure calculation. The Brillouin zone integration in the limit of infinite relaxation time so far converges faster with respect to the number of $\boldsymbol{k}$-points. Hence, we may conclude that the discussed approach is in fact the method of choice if the number of $\boldsymbol{k}$-points is limited in the band structure calculation. However, it is only applicable if the system satisfies the symmetry assumptions. Heading towards the calculation of optical conductivities of more complex systems, the direct evaluation of the Kubo formula Eq. (1) will become more important.

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