Interpolation between spectra satisfying sum rules

Hans-Christian Weissker,[1,2,*] Ralf Hambach,[1,2] Valerio Olevano,[2,3] and Lucia Reining[1,2]
[1]*Laboratoire des Solides Irradiés, Ecole Polytechnique, CNRS, CEA/DSM, F-91128 Palaiseau, France*
[2]*European Theoretical Spectroscopy Facility (ETSF)*
[3]*Institut Néel, UPR 2940, CNRS & UJF, F-38042 Grenoble, France*
(Received 17 January 2008; revised manuscript received 11 November 2008; published 5 March 2009)

A method is presented which is able to interpolate between spectra depending parametrically on one variable and obeying a sum rule. This enables the description of experiments with a finite resolution in that parameter because integrals over certain parameter ranges are easily obtained, as for instance in the case of inelastic x-ray scattering with finite resolution in momentum transfer. Beyond the sum rule, the method does not use further assumptions about the physics of the system. It is applicable to a wide range of spectra as for instance the dynamic structure factor or the dielectric function for different moduli or directions of momentum transfer, absorption spectra for different alloy compositions or for a range of nanocrystal sizes, probability distributions, etc. The method is expected to be useful not only in the simulation of experimental spectra but also in calculations where the determination of certain spectral quantities is numerically cumbersome. A code carrying out the interpolation is provided.

DOI: 10.1103/PhysRevB.79.094102 PACS number(s): 71.15.Dx, 77.22.−d, 78.20.Bh, 31.15.B−

I. INTRODUCTION

Many spectroscopic experiments investigate spectra that depend parametrically on some quantity, for instance, on the momentum transfer **q** in inelastic x-ray scattering (IXS) (Ref. 1) or in electron energy-loss (EEL) spectroscopy,[2] and which obey certain sum rules. As the **q** resolution of any experiment is necessarily finite, the spectra obtained represent an average over the desired quantity within the **q** range determined by the resolution.[3]

Therefore, in general, each experimental spectrum is an integral of the form

$$f_{\mathrm{expt}}(q,\omega) = \int dq' f(q',\omega) g(q-q'), \tag{1}$$

with some weight function $g(q)$. The calculation of the measured spectrum requires, therefore, the knowledge of the q dependence of the functions $f(q,\omega)$ or at least their values on a very fine q grid in order to calculate the integral Eq. (1) numerically.

A similar situation occurs for instance when the absorption spectrum of a sample of nanocrystals is measured. In this case, the size of the nanocrystals takes the role of the parameter q, and the measured spectrum represents an integral over the size-dependent spectra of the nanocrystals in the sample, weighted by the size distribution.

On the other hand, calculations, today often within the framework of *ab initio* approaches,[4] usually calculate the spectra for one fixed **q** or one fixed size of a nanocrystal at a time. The state-of-the-art calculation of those spectra is numerically often very demanding. For instance, the calculation of one high-precision energy-loss spectrum in silicon for large momentum transfers using time-dependent density-functional theory (TDDFT) in the adiabatic local-density approximation (TDLDA) (Ref. 5) takes about 7 h on a NEC SX8. The calculation of spectra using more sophisticated theoretical approaches, as for instance self-consistent *GW* calculations, for more complicated materials, as for instance $V0_2$,[6] can easily take 1 or 2 orders of magnitude more time. On the other hand, the calculation of an absorption spectrum of a germanium nanocrystal of 83 atoms using TDLDA in time evolution with the octopus code[7] takes about 1 day on 32 processors once the ground-state calculation is done. Moreover, the standard calculation of response functions in solids is often restricted to certain momentum transfers **q** by the discrete sampling of the Brillouin-zone.

The calculation of many curves on a fine parameter mesh is therefore often not practicable. Hence the question arises as to how curves for intermediate parameter values can be obtained. A straightforward average, for instance, between two calculated curves does not give a good result. In particular changes in a peak's position are not accounted for. But even in the case of peaks centered at the same point, the average does not yield the correct result expected for intermediate parameter values.

Considerations about changing spectra are, therefore, often based on a fitting of separate (Gaussian, Lorentzian, etc.) contributions to the spectra, the fitting parameters of which may afterward be interpolated. However, this amounts to assumptions about the physical nature of the spectra. In particular for more complicated spectra, the complexity can be prohibitive for the finding of reasonable fits. On the other hand, approaches starting from the physical nature of the system under study can be devised which treat interpolation on the basis of the physical description.[8] These methods are necessarily restricted to a given system.

Any procedure providing interpolation must be able to shift peaks and spectral weight. This would obviously be accomplished by an interpolation along the abscissa (energy axis for spectra). However, this problem does not lend itself to a simple solution because, while for a given energy ω the spectrum $f(q=\mathrm{const},\omega)$ is unique, for a given value of $f(q=\mathrm{const},\omega)$ there is no unique value of ω associated. This bars any simple attempts of interpolation. In other words, the problem is that the spectra are not monotonous.

On the other hand, a monotonous function to be interpolated can be the solution of this problem. Moreover, many

spectra obey certain sum rules. We combine these two points. The proposed method solves the problem by calculating a distribution-function-like monotonously increasing function[9] integrating the data, used thereafter for an interpolation in the ω direction. (Due to the linearity of the integration, "normal" interpolation along the ordinate would just reproduce the averages.)

The present paper describes the idea and the procedure in Sec. II while in Sec. III the behavior in certain limiting situation is discussed using model spectra. In Sec. IV, examples are presented for the method applied to real data.

II. IDEA AND METHOD

We consider curves which obey a sum rule of the form

$$\int_0^\infty f(q,\omega)d\omega = \text{const}, \tag{2}$$

where $f(q,\omega)$ are functions of ω which depend parametrically on q. If $\mathbf{q}$ is a vector, as in the case of the momentum transfer, we choose q as the modulus of $\mathbf{q}$ in a given direction. However, the method should easily be extendable to three-dimensional $\mathbf{q}$ space. The curves to be interpolated have to be positive definite. Sum rules of the form

$$\int_0^\infty f(q,\omega)d\omega = C(q), \tag{3}$$

where $C(q)$ depends only on q, can easily be brought into form (2) using the dependence $C(q)$.

Examples are the dynamic structure factor $S(\mathbf{q},\omega)$ in IXS with the sum rule (a.u.) (Ref. 10)

$$\int_0^\infty S(\mathbf{q},\omega)\omega\, d\omega = q^2/2, \tag{4}$$

which depends parametrically on the momentum transfer $\mathbf{q}$, or the electron energy-loss function $-\text{Im}\ \varepsilon^{-1}(\mathbf{q},\omega)$ which follows the same sum rule but integrates to a $\mathbf{q}$-independent constant. Another example is the imaginary part of the dielectric function for, e.g., changing size of nanocrystals or changing composition of alloys. It obeys the generalized oscillator-strength sum rule[11]

$$\int_0^\infty \omega\ \text{Im}\ \varepsilon(\omega)d\omega = \frac{\pi}{2}(\omega_p^{\text{eff}})^2, \tag{5}$$

and the screening sum rule

$$\int_0^\infty \frac{1}{\omega}\text{Im}\ \varepsilon(\omega)d\omega = \frac{\pi}{2}(\varepsilon_\infty - 1), \tag{6}$$

where ω_p^{eff} is the effective plasma frequency and $\varepsilon_\infty = \text{Re}\ \varepsilon(\omega=0)$ is the (high-frequency) dielectric constant.[11] In the case of, for example, nanocrystals of different sizes, the radius r can be used for the general parameter q. In the sum rule, we have now $\varepsilon_\infty(r)$. Knowing $\varepsilon_\infty(r)$ for the radii of the inputs, $\varepsilon_\infty(r)$ can then itself be interpolated in order to obtain $\varepsilon_\infty(r)$ for the radii to be calculated using the present method.

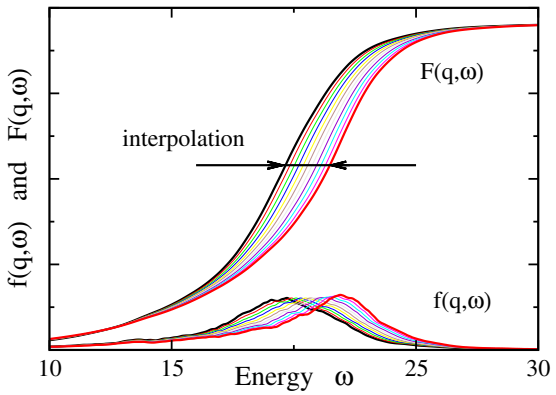

FIG. 1. (Color online) Illustration of the method—the integrals Eq. (7) for two different input curves (thick lines) and indication of the interpolation direction. Also shown are a number of interpolated curves (linearly between the two inputs) for the integrals and the resulting spectra.

The following procedure accomplishes the interpolation for arbitrary intermediate values of the parameter q:

(i) Using the factors $C(q)$ of the sum rules, the curves are rescaled such that the sum rule of the form Eq. (2) is recovered.

(ii) Then the integral

$$F(q,\omega) = \int_0^\omega f(q,\omega')d\omega', \tag{7}$$

which for $\omega\to\infty$ gives the sum rule is calculated for all ω. This results in monotonously increasing functions $F(q,\omega)$ which for $\omega\to\infty$ attain all the same value, i.e., which all span the same range along the ordinate (cf. Fig. 1).[12]

(iii) The integrals are then interpolated along the ω direction [i.e., for each fixed $F,\omega(F,q)$ is interpolated along the parameter q] corresponding to the desired q values as indicated in Fig. 1. This interpolation shifts spectral weight and in this way takes care of the shift of peak positions. The interpolation is done pointwise as explained below in Sec. II A, using linear or higher-order polynomial interpolations or splines.

(iv) The interpolated curves are then differentiated in order to get the desired spectra. Depending on the sum rule, the inverse scaling according to the parameters of the sum rule $C^{-1}(q)$ must be applied for the respective q values of the interpolated functions.

With the same approach, *extrapolation* to q values beyond the limits given by the input curves is likewise possible.

We note that the problem can also be stated in terms of a function f of two variables q and ω which is known along lines of fixed q (Fig. 2). The task consists in providing $f(q,\omega)$ over the whole (q,ω) plane. We mention that the standard approaches for interpolation of two-dimensional (n-dimensional) functions do not yield the physically meaningful results obtained by the present method.

Within the picture of the surface over the (q,ω) plane, the method that we present can be interpreted as follows. For the integrals $F(q=\text{const.},\omega)$ of the input spectra, the intersections with the plane $F(q,\omega)=\text{const.}$ parallel to the (q,ω)

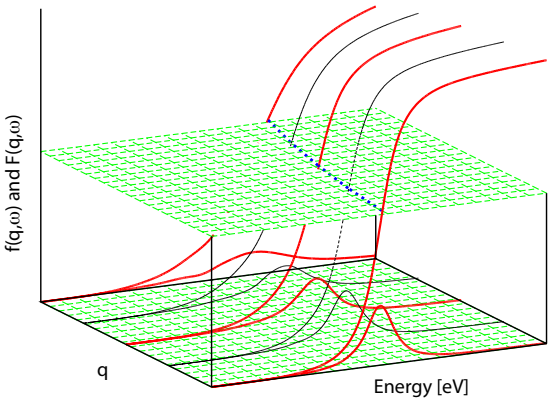

FIG. 2. (Color online) Interpretation of the interpolation method within the picture of a function f of the two variables q and ω. Shown are the input spectra $f(q,\omega)$ for three values of q (thick red lines), as well as two interpolated spectra (thin black lines), along with the integrals given with the corresponding line types. The blue dotted line shows the (here quadratic) interpolation between the intersections of the integrals with a plane $F(q,\omega)$=const. within this plane. The curves used for this schematic figure are the EEL spectra shown in Fig. 10. We show as $f(q,\omega)$ the spectra multiplied by ω, as well as the integrals $F(q,\omega)$ of this quantity. The intermediate spectra have been calculated using the present method.

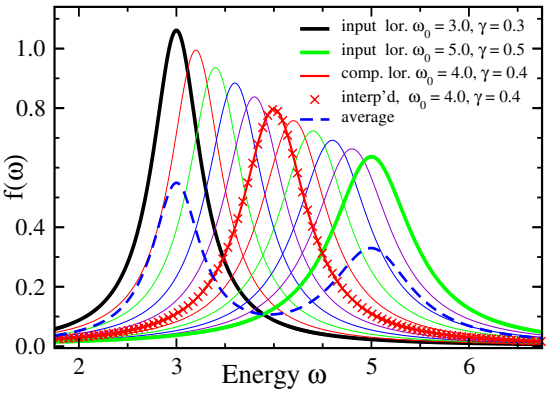

FIG. 3. (Color online) Interpolation (linear) between input Lorentz curves (thick black and green) with parameter γ and center ω_0 as indicated. The interpolated curve in the middle coincides precisely with the Lorentz curve (thick red) calculated for comparison with these intermediate parameters. (The interpolated result is given by the crosses to make the equality apparent.) Also shown are a number of interpolated curves (thin lines). The simple average of the input curves (blue dashed) gives a double-peak structure far from the desired result.

plane are interpolated within this plane. It is here where we can choose linear, polynomial, etc. interpolation, depending on the situation and inputs. This is shown in Fig. 2 for one plane. Carrying out the interpolation for all planes $F(q,\omega)$ =const. intersected by the integrals, the values of the integrals of intermediate q values are found which then yield the interpolated spectra.

This three-dimensional picture is useful for the understanding of the method we present. The latter yields the interpolated spectra without the complication of three-dimensional space. Moreover, the method can easily be extended to three-dimensional parameter space **q**, where the three-dimensional representation looses its highly intuitive character.

A. Implementation

A code using the procedure described here is available.[13] The idea is implemented as follows: after a possible rescaling, the data are first integrated. The interpolation of the integrals along the energy direction is done by exchanging abscissa and ordinate and, thereafter, interpolating between the integrals. This interpolation is done pointwise, using either linear or quadratic interpolation (first-order or second-order Taylor expressions with finite difference expressions for the coefficients), or cubic or Akima splines. The interpolation is implemented using the gnu scientific library (gsl).[14] The exchange of abscissa and ordinate corresponds to an inversion $F(q,\omega)\rightarrow\omega(q,F)$. For the interpolated integrals we exchange again abscissa and ordinate which gives the integrals that are finally differentiated to obtain the desired spectra. These might have to be rescaled according to the scaling at the beginning. For a typical spectrum, the program runs a few seconds.

For the moment, the method works along one direction of q, i.e., for a scalar parameter. The generalization to three-dimensional **q** space is not expected to present any difficulty.

III. ACTION ON MODEL INPUTS

A. Sum of peaks with equal spectral weight

The spectra used here have been created as sums of peaks (Gauss, Lorentz) in such a way that each single peak gives the same spectral weight to the sum-rule integral of Eq. (2), i.e., for every peak the integral $\int_a^b f(\omega)d\omega$ gives the same value, with a and b being the lower and the upper limits of the peaks. In order to investigate the behavior of the method for a single peak which changes its position and its broadening, we show different Lorentz curves whose parameters are changed linearly as shown in Fig. 3. This corresponds to a linear parametrization of the parameters γ and ω_0 of the Lorentz function $\gamma/\pi[(\omega-\omega_0)^2+\gamma^2]$ with respect to some parameter q. Throughout the paper we use a unitless energy scale for the model curves. In all the figures, *input* refers to the curves used as input for the interpolation procedure, *compare* refers to curves created for comparison using the corresponding parameters, and *interpolated* refers to the result of the interpolation.

The curve created with the intermediate parameters, and the one using the present interpolation procedure coincide with high precision. The method thus reproduces the change in both peak position and broadening. Problems might arise when the integrals of Eq. (7) cross, which is discussed below.

As an illustration of the fact that a simple average does not yield a reasonable approximation of intermediate curves, the average of the two input curves, a double-peak structure, is also shown in Fig. 3.

B. Crossing of peaks

Now we look at the situation where we have two peaks of different broadening at two energies. We face here a problem

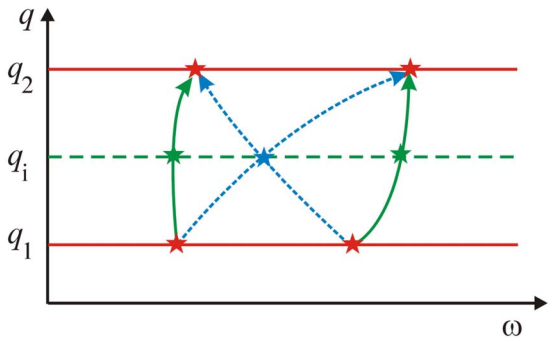

FIG. 4. (Color online) Schematic of the ω-q plane showing the two possibilities to interpolate between two spectra exhibiting two peaks each, indicated by the asterisks (red). On the one hand, the peaks can move without crossing, as indicated by the green solid arrows. At all spectra for intermediate q, there will be two peaks. On the other hand, the peaks may cross according to the blue dotted arrows.

which always arises in the interpolation between two different functions at two points—there are two ways to connect them. In one case, the two functions cross while in the other they do not.

This crossing has its equivalent in the exchange of the peak positions as illustrated in the schematic in Fig. 4. The situation of different q with two peaks each is illustrated by the inputs (thick black and dashed green) of Fig. 5 which are each a sum of two Lorentz peaks with different widths. Two situations are now possible for the interpolation: either the two peaks exchange their position, or they stay at their place and each one goes through intermediate values of the width. The two situations correspond to two different parametrizations of $f(q,\omega)$: In the non-crossing case (Fig. 5), the broadenings $\gamma(q)$ change linearly with q while the centers ω_0 are kept constant. In the crossing case, the positions $\omega_0(q)$ change linearly with q while the widths γ remain constant.

The non-crossing behavior (Fig. 5) is what the method (linear interpolation between two inputs) does when used

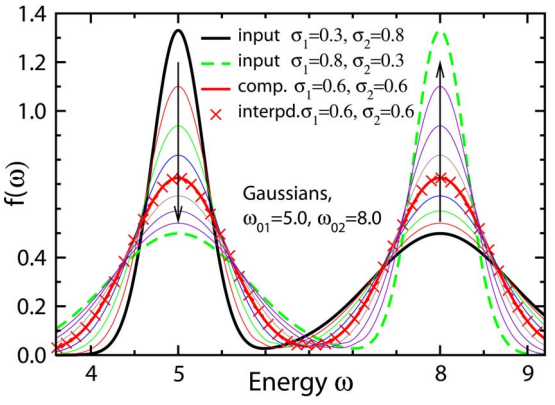

FIG. 5. (Color online) Spectrum made of two peaks which change their broadening. The dashed green and the thick black lines are the inputs, and the thin lines are the curves obtained for intermediate q using the interpolation method (linear interpolation). σ_i and ω_{0i} refer to the first and the second peaks, respectively. The arrows are a guide for the eyes to clarify the direction of the change in the peaks (black→green dashed).

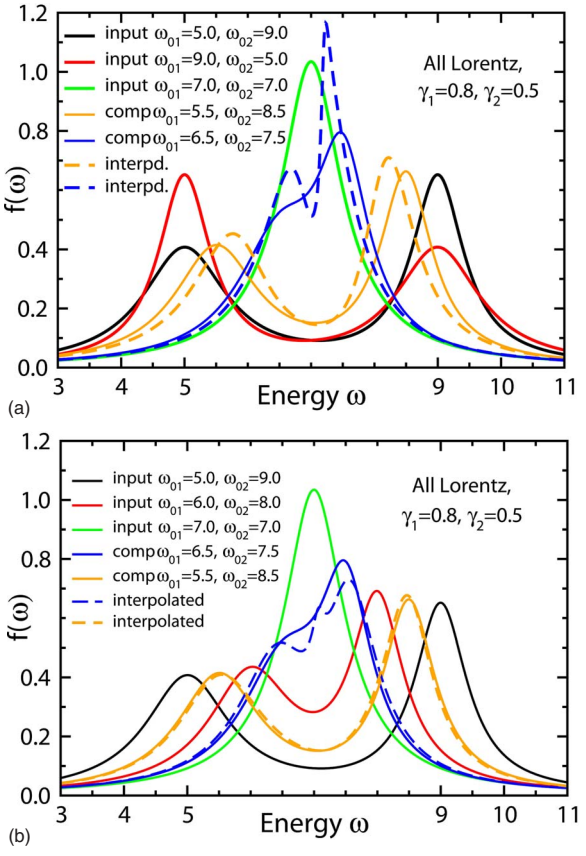

FIG. 6. (Color online) Same situation as in Fig. 5 for the two outer input spectra (thick black and red) but with a third input spectrum (thick green) created using the intermediate parameters which forces a crossing of the peaks. σ_i and ω_{0i} refer to the first peak and the second peak, respectively. Panel (a): second-order polynomial interpolation using the two outer (black and red) and the middle (green) peaks as inputs. The interpolated curves (dashed) for two q are compared with the result created for these values (solid). Panel (b): second-order polynomial interpolation between the black and the middle green inputs as in (a), with an additional input between the two (red).

without any further constraint. However, by providing a third input curve at the intermediate position, the method can also be forced to simulate the exchange of peak positions. This is shown in Fig. 6(a). Due to the parametrization, the middle input curve is the sum of two peaks with the same ω_0 which results in only one peak. The interpolated result shows that the two peaks move roughly in the right way but the interpolated curves show rather large deviations from the curve calculated directly using the interpolated parameters.

A possible solution, shown in Fig. 6(b), is to use another input curve between one of the outer and the middle curve. The result, reasonable for a limiting situation such as the crossing, provides now interpolated curves which differ much less from the spectra calculated for comparison.

The deviations here appear to be a shortcoming of the interpolation under the constraint given by the curve at intermediate position. This sort of shortcoming appears to be connected with the situation where the integrals of Eq. (7) cross,

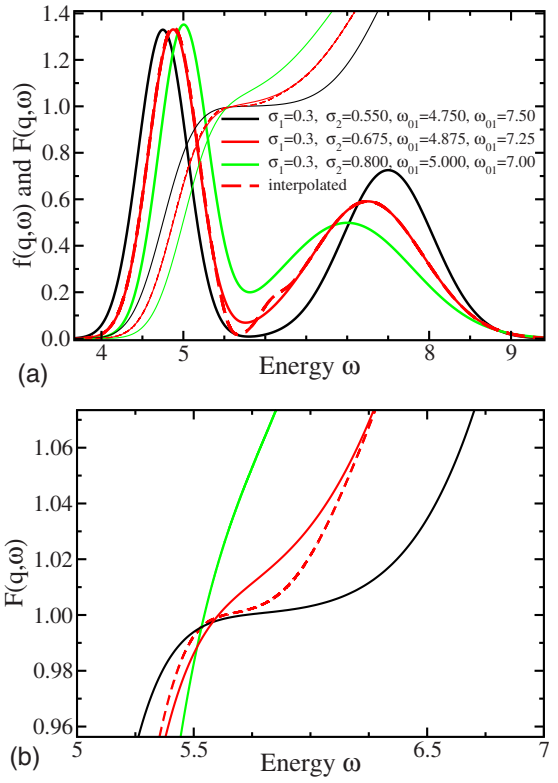

FIG. 7. (Color online) (a) Input spectra composed of two Gaussians with parameters as indicated, σ_i and ω_{0i} referring to the first peak and the second peak, respectively. The two outer curves (black and green) are taken as input for the (linear) interpolation, and the result (red dashed) is compared with a curve created for comparison using the corresponding parameters (red solid). Also shown are the integrals according to Eq. (7) showing the crossing (corresponding colors, thin lines). (b) Blowup of the region of the crossing showing the integrals of panel (a). Colors as in (a).

discussed below. Moreover, the result in this case depends rather strongly on the interpolation chosen (linear or quadratic Taylor, splines, etc.).

C. Crossings of the integrals

Figure 7 shows a situation which presents a problem. The input curves are two double-peak structures of two Gaussians each with parameter values as indicated in the figure. In this case, the integrals $F(q,\omega)$ of Eq. (7) cross. Around this point, the linear interpolation between the integrals does not produce a good interpolated curve. This is shown in Fig. 7 where between ω=5.5 and 7.5 a serious deviation of the interpolated curve is found compared to the curve created with the corresponding parameters. This can be seen in the integrals as well. In Fig. 7(b) the enlarged region of the crossing demonstrates that the interpolation between the integrals, linear in this case, does not represent the situation well.

In general, the crossings of the integrals give rise to problems in certain situations. Concerned are situations such as the one shown in Fig. 7(b) where the crossing occurs under a rather large angle and one of the integrals is almost horizontal after the crossing. A crossing under a small angle does not create these problems.

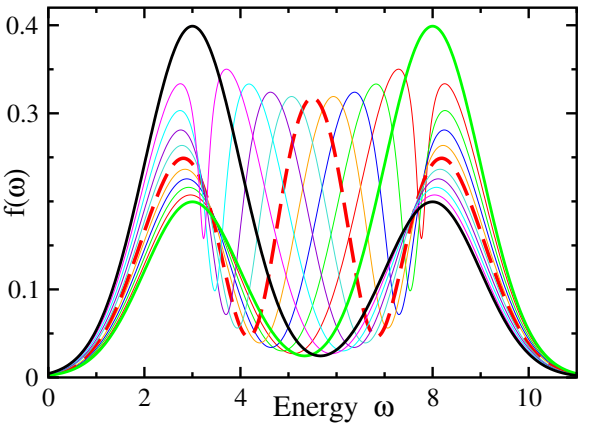

FIG. 8. (Color online) Input spectra (thick green and black) as sums of peaks which give different weights to the sum rule integrals. While the two parts of the peaks which do have the same weight behave roughly like in Fig. 5, a third peak carries the weight from one side to the other (red dashed).

Technically, the treatment of such points depends strongly on the interpolation method used. In particular in the case of quadratic Taylor interpolation using three input integrals, the crossings present a problem when the integrals of all three inputs cross at roughly the same point. In these cases, the result can be even more distorted than the example in Fig. 7, with a singularity-like peak added to the interpolated curve. This happens because the interpolation is done pointwise without a stipulation that the output curves have the same monotony as the inputs. However, inspection of the results reveals very quickly the few points where this problem occurs. In practice, these points should be treated with the most convenient interpolation method (probably pointwise linear between the inputs). The use of more inputs to interpolate between will also solve the largest part of the problem.

Another way of approaching this problem is by adding a constant to the input curves. In this way, the angle under which the integrals cross is reduced, and the almost horizontal part of the lowest integral in Fig. 7(b) has a larger slope. The result of this procedure is that the deviations of the interpolated results from the "true" curves occur in different places. The rest of the curves is only slightly changed. (Tests with input spectra that do not exhibit problems such as the crossing indicate that the use of such a small offset leads to only very small changes.) Therefore, adding the shift may help in obtaining good results by doing the interpolation for different parts of the spectrum one after the other.

D. Peaks that do not separately fulfill the sum rules

Now we consider the situation when the model spectrum is composed of peaks which do not each contribute the same spectral weight to the sum rule. Figure 8 shows model spectra that contain two peaks which change their widths such as in the case before, but now one of them gives twice as much weight to the integral as the other. The procedure acts in such a way as if the larger peak was composed of two peaks: one

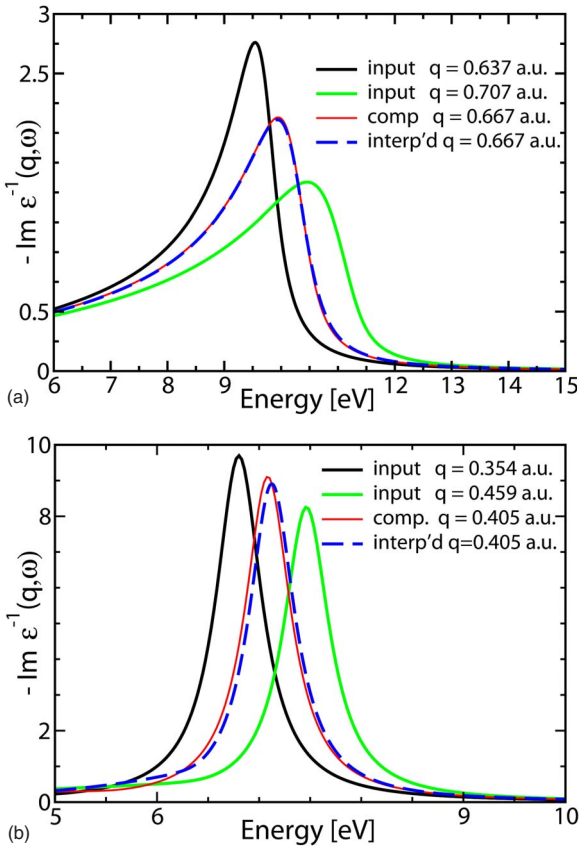

FIG. 9. (Color online) Interpolation (linear) between the loss function of the homogeneous electron gas for r_s=4.0 and momentum transfers as indicated.

which is left in place such as the one above in Fig. 5 while another peak moves between the two to "carry the spectral weight" from the left to the right. Inspection of the expected result, as in the case of two peaks exchanging their position above, must determine if this is the desired behavior. The hypothetical situation of two separated peaks which exchange spectral weight—untypical for spectroscopy—is hence not described by the present procedure.

IV. REALISTIC EXAMPLES

A. Electron energy-loss function of the homogeneous electron gas

We show here the application of the procedure to realistic physical spectra. We first treat the electron energy-loss function of the homogeneous electron gas. In Fig. 9 we show the linear interpolation between two relatively small as well as between two larger momentum transfers q using the sum rule Eq. (4). As the system is isotropic, the modulus of the momentum transfer is sufficient as parameter. The agreement is good, especially for larger momentum transfer.

B. Electron energy-loss function of silicon

As a second example, Fig. 10 shows a series of EEL spectra $-\mathrm{Im}\,\varepsilon^{-1}(\mathbf{q},\omega)$ of Si calculated in the TDLDA.[5] In this case, the parameter q of Eq. (4) is the modulus of $\mathbf{q}$, and the latter is parallel to the [111] direction. The interpolation is carried out over a rather large range of $\mathbf{q}$ (half the extension of the Brillouin zone in this direction), and the interpolated curves for intermediate values of $\mathbf{q}$ are compared to curves which have been calculated directly. The agreement and, therefore, the quality of the interpolation are very good.

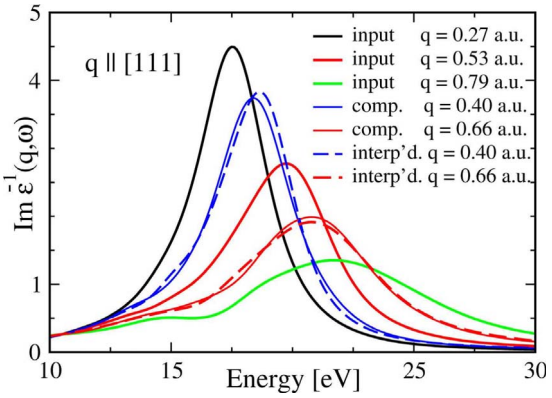

FIG. 10. (Color online) Interpolation (second-order polynomial) between loss spectra of bulk silicon calculated within TDLDA with inclusion of lifetimes (Ref. 5) for three $\mathbf{q}$ values (thick black, red, and green lines). The intermediate calculations (thin red and blue) are compared with the interpolated curves (dashed red and blue).

This example shows that the spectra do not have to be known over an energy range large enough to *satisfy* the sum rule. It is sufficient if the spectra are normalized and the sum rule is known. This situation appears often in calculations where the normalization factors are correct but the energy range to be calculated is restricted because the calculations are numerically heavy.

C. Dielectric function of nanocrystals and alloys

Another application of the method can be the calculation of spectra for a size distribution of nanocrystals in a sample.

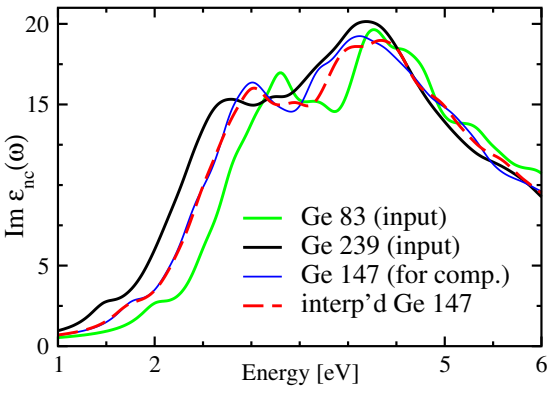

FIG. 11. (Color online) Interpolation (linear) between the imaginary part of the normalized dielectric function of Ge nanocrystals (Ref. 15) for two sizes, Ge 83 and Ge 239 (solid lines as indicated). The numbers indicate the number of Ge atoms in the nanocrystal. The calculation of the intermediate size Ge 147 (thin blue) is compared with the interpolated curve (dashed red) for the same size.

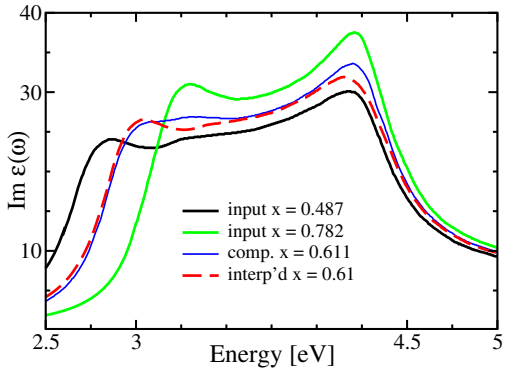

FIG. 12. (Color online) Interpolation (linear) between the imaginary parts of the dielectric function of the $Ge_{1-x}Si_x$ bulk alloy of different composition x. The measurements used for input and comparison are from Ref. 16.

In order to calculate the spectra of the size distribution, one needs the spectra for all sizes of nanocrystals intermediate to a given set of calculations. The spectra shown in Fig. 11 are the imaginary part of the normalized dielectric function of nanocrystals (cf. Fig. 6 of Ref. 15) for three Ge nanocrystals. The parameter which the sum rule depends on is now the radius r of the nanocrystal.

We use the screening sum rule [Eq. (6)] with the values of $\varepsilon_\infty(r)$ taken from the same calculations. Equation (6) has now the form of Eq. (3), with $f(q,\omega)\to \mathrm{Im}\,\varepsilon(r,\omega)/\omega$ and $C(q)\to\pi[\varepsilon_\infty(r)-1]/2$. The $\varepsilon_\infty(r)$ for the interpolated curve has been linearly interpolated between the values corresponding to the two inputs. The result is evidently satisfactory, in particular when the interpolated curves are used for an averaging over a size distribution.

The last example that we present (Fig. 12) shows again the dielectric function, this time for the $Ge_{1-x}Si_x$ bulk alloy with different compositions x. The experimental values are taken from Ref. 16. The $\varepsilon_\infty(x)$ is again interpolated linearly, as $\varepsilon_\infty(x)=16.2-4.5x$.

V. CONCLUSION

In conclusion, we present a method which interpolates between spectra belonging to different values of some parameter, and which obey a sum rule. No assumptions about the underlying physics have to be made beyond the validity of the sum rule and the positive definiteness of the spectra. For single Gaussian or Lorentzian peaks, the dependence on the values of their parameters is precisely reproduced. Problems arise in cases where the integrals cross under large angles, in particular in cases where two peaks exchange their positions. If the spectra are made of peaks which do not all give the same weight to the sum rules, the procedure treats them roughly as a sum of such. The procedure is applicable to a wide range of spectra. A code which performs the interpolation is made available.

ACKNOWLEDGMENTS

We thank M. Cazzaniga for providing help with the HEG response function. The work was supported by the European Union through the NANOQUANTA Network of Excellence Contract No. NMP4-CT-2004-500198, the eI3 Contract No. (INFRA-2007-211956), and the Marie-Curie Training Site Contract No. HPMT-CT-2001-00368. Computer time was granted by IDRIS Project No. 020544 on the NEC SX5. H.-C.W. acknowledges support from the European Union through the individual Marie Curie Intra-European Grant No. MEIF-CT-2005-025067.

*Present address: Laboratorie de Physique des Interfaces et Couches Minces, Ecole Polytechnique, F-91128 Palaiseau Cedex, France. hansi.weissker@polytechnique.fr

[1] W. Schülke, in *Handbook of Synchrotron Radiation*, edited by G. Brown and D. E. Moncton (Elsevier Science, New York, 1991), Vol. 3, Chap. 15.

[2] R. Brydson, *Electron Energy-Loss Spectroscopy* (Garland Science, London, 2001).

[3] Here and in the following we restrict our treatment to a one-dimensional parameter. In the case of the momentum transfer, this could be either the direction of **q** or its modulus along a given direction. A generalization to three-dimensional parameters such as **q** itself is straightforward.

[4] G. Onida, L. Reining, and A. Rubio, Rev. Mod. Phys. **74**, 601 (2002).

[5] H.-Ch. Weissker, Jorge Serrano, S. Huotari, F. Bruneval, F. Sottile, G. Monaco, M. Krisch, V. Olevano, and L. Reining, Phys. Rev. Lett. **97**, 237602 (2006).

[6] M. Gatti, F. Bruneval, V. Olevano, and L. Reining, Phys. Rev. Lett. **99**, 266402 (2007).

[7] A. Castro, H. Appel, Micael Oliveira, C. A. Rozzi, X. Andrade, F. Lorenzen, M. A. L. Marques, E. K. U. Gross, and A. Rubio, Phys. Status Solidi B **243**, 2465 (2006).

[8] J. J. Max and C. Chapados, Appl. Spectrosc. **53**, 1601 (1999).

[9] "Distribution" is meant in the sense of the distribution functions of probability distributions.

[10] See for instance J. Z. Tischler, B. C. Larson, P. Zschack, A. Fleszar, and A. G. Eguiluz, Phys. Status Solidi B **237**, 280 (2003).

[11] See for instance B. Adolph, V. I. Gavrilenko, K. Tenelsen, F. Bechstedt, and R. Del Sole, Phys. Rev. B **53**, 9797 (1996).

[12] In some cases, the scaling has a clear physical interpretation, e.g., in the case of the dynamic structure factor $S(\mathbf{q},\omega)$ of Eq. (4). In this case, the scaling creates functions which are proportional to the electron energy-loss function $-\mathrm{Im}\,\varepsilon^{-1}(\mathbf{q},\omega)$, the sum rule of which is not q dependent. The interpolation is hence done between the loss functions for different **q** before the $S(\mathbf{q},\omega)$ is regained using the inverse scaling.

[13] A code which carries out the interpolation can be found at http://etsf.polytechnique.fr/Software/Interpolation

[14] M. Galassi, J. Davis, J. Theiler, B. Gough, G. Jungman, M. Booth, and F. Rossi, *GNU Scientific Library Reference Manual*, 2nd ed. (Network Theory Ltd., 2003); www.gnu.org/software/gsl

[15] H.-Ch. Weissker, J. Furthmüller, and F. Bechstedt, Phys. Rev. B **65**, 155328 (2002).

[16] J. Humlicek, M. Garriga, M. I. Alonso, and M. Cardona, J. Appl. Phys. **65**, 2827 (1989).