Accurate and efficient implementation of the von Neumann representation for laser pulses with discrete and finite spectra

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Abstract. We recently introduced the von Neumann picture, a joint time–frequency representation, for describing ultrashort laser pulses. The method exploits a discrete phase-space lattice of nonorthogonal Gaussians to represent the pulses; an arbitrary pulse shape can be represented on this lattice in a one-to-one manner. Although the representation was originally defined for signals with an infinite continuous spectrum, it can be adapted to signals with discrete and finite spectrum with great computational savings, provided that discretization and truncation effects are handled with care. In this paper, we present three methods that avoid loss of accuracy due to these effects. The approach has immediate application to the representation and manipulation of femtosecond laser pulses produced by a liquid-crystal mask with a discrete and finite number of pixels.

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1. Introduction

The field of femtochemistry, which came into being about 20 years ago, has flourished, with probably several hundred laboratories around the world working in this general area \[1\]–\[4\]. Since the beginning of the femtochemistry era (and to some extent even before) it was realized that shaped femtosecond pulses could have profound effects on photochemical reactions \[5\]–\[10\]. Various techniques have been developed to optimize the shape of femtosecond pulses, specifically techniques from the calculus of variations and optimal control theory \[5\]–\[7\], \[9\]. In recent years, many groups have exploited evolutionary algorithms to design optimized pulses using adaptive control \[10\]–\[17\]. Femtosecond pulse sequences have also proven useful in multidimensional optical and IR spectroscopy \[18\]–\[20\].

One of the main challenges facing the field of femtosecond pulse shaping is interpreting the mechanism of shaped pulses. A first step toward interpretation is to visualize the pulse in a way that reveals its key properties. Pulses are normally depicted in either the time or the frequency domain; although each of these representations is obviously valuable, there are aspects of the pulse that are obscured if the pulse is visualized in time or frequency alone. Is it possible to view the properties of the pulse in time and frequency simultaneously? The answer is yes: by analogy with the use of a position-momentum phase space to visualize wavefunctions in quantum mechanics \[21, 22\], pulses may be represented in a time–frequency phase space. In a series of recent papers \[23, 24\] we have focused on one such phase space picture known as the von Neumann representation. It is related to, but somewhat less familiar than the Wigner and Husimi phase-space representations. In particular, the von Neumann representation exploits a discrete phase-space lattice of nonorthogonal Gaussians as a basis to represent an arbitrary pulse. This basis is complete but not overcomplete \[25\], and therefore provides a one-to-one mapping from either the time or the frequency representation of the pulse.

It is intuitively obvious that if a function is of finite range in both conjugate variables it should be possible to truncate the von Neumann lattice to match the range spanned by the functions. In our previous work, we used such a truncation of the von Neumann lattice without systematically analyzing the effects of truncation; although the overall reconstruction of pulses after back-transforming from the von Neumann representation was excellent, small distortions...
were observed [23]. In this paper, we analyze the origin of these distortions and then discuss several possible solutions to remove them. Additional motivation for the present work comes from the fact that shaped femtosecond pulses are normally produced using a liquid crystal display (LCD) mask, corresponding to a finite and discrete set of frequencies. One suspects that since the truncated von Neumann lattice is also finite and discrete there should be a one-to-one mapping from the discrete representation in frequency to the discrete time–frequency plane. One of our approaches to dealing with the truncation error indeed corresponds to such a one-to-one mapping from one discrete space to the other.

In the final stages of writing this manuscript, we learned that the use of the von Neumann lattice in the time–frequency representation has a long history in signal analysis, tracing back to Gabor in 1946 [26]. It seems that the use of the method was held back by the difficulty in accurately computing the Gabor coefficients (coefficients of the Gaussians on a lattice). In the 1980s, Bastiaans made two seminal contributions. Firstly, he introduced the biorthogonal function for the Gaussian window (this is a linear algebraic formalism that essentially incorporates the inverse overlap matrix into the coefficients of the nonorthogonal basis functions) [27]. Secondly, he showed that the biorthogonal formalism allows a simple formal generalization to window functions other than Gaussians, thereby providing the basis of modern Gabor theory [28]. In the 1990s several studies were performed on the discretization of the Gabor transform and its ancillary functions by periodizing and sampling. The work of Orr [29] puts earlier work on this subject by Auslander et al [30, 31] and Wexler and Raz [32], into a single, unified framework. Orr has also derived a sampling theorem, essentially a composite Nyquist rate, for finite discrete Gabor transforms, an issue related to our work on the connection between the truncated von Neumann lattice and the truncated discrete Fourier transform [33].

The organization of the paper is as follows. In section 2, we review the von Neumann formalism. In section 3, we illustrate the numerical error that can be incurred using a truncated von Neumann lattice. In section 4, we present three approaches to overcome these errors and discuss the advantages and disadvantages of each. Section 5 concludes.

2. von Neumann formalism

Before starting a detailed analysis of the effect of truncation of the von Neumann lattice, we review the basic properties of the von Neumann representation [23].

The von Neumann representation for ultrashort laser pulses corresponds to a mapping of the electric field

$$\varepsilon (\omega) = |\varepsilon (\omega)| \exp [-i\Phi (\omega)]$$

in the frequency domain onto a joint time–frequency domain, the so-called von Neumann plane. The method exploits a discrete lattice of complex Gaussians to represent the pulses; an arbitrary pulse shape is represented in terms of its values at each of the lattice points. The complex Gaussians have the form [25, 34, 35]

$$\tilde{a}_{\omega n} t_m (\omega) = \left( \frac{2\alpha}{\pi} \right)^{1/4} \exp \left[ -\alpha (\omega - \omega_n)^2 - it_m (\omega - \omega_n) \right].$$

Note that these basis functions remain Gaussian when transformed to the time domain. The Gaussians are centered at a set of equally spaced points ($\omega_n, t_m$) in the joint time–frequency
domain. The Gaussians at different lattice sites are nonorthogonal. They provide a complete but not overcomplete basis provided that the lattice points are placed at the vertices of rectangles with area $2\pi$ and $\alpha$ is chosen to be $\pi/(\Omega \delta \omega)$, where $\Omega$ is the frequency range of the considered discrete signal and $\delta \omega$ the corresponding stepsize.

Using this basis set, the electric field can be written as

$$\varepsilon (\omega) = \sum_{n,m} \tilde{Q}_{\omega n t m} \tilde{\alpha}_{\omega n t m}(\omega),$$

where the complex valued von Neumann coefficients $\tilde{Q}_{\omega n t m}$ are evaluated as

$$\tilde{Q}_{\omega n t m} = \sum_{i,j} S^{-1}_{(n,m)(i,j)} \langle \alpha_{\omega i t j} | \varepsilon \rangle.$$

The overlap matrix

$$S_{(n,m),(i,j)} = \langle \alpha_{\omega i t j} | \alpha_{\omega n t m} \rangle$$

takes into account the nonorthogonality of the von Neumann basis functions. An analytic expression for (5) can be derived by evaluating the resulting Gaussian integrals [23].

In the following, we call the overlap between the electric field and the von Neumann basis functions,

$$Q_{\omega n t m} = \langle \alpha_{\omega n t m} | \varepsilon \rangle = \int \tilde{\alpha}_{\omega n t m}^* (\omega) \varepsilon (\omega) \, d\omega,$$

the ‘complex Husimi’ representation because its absolute value squared is equivalent to the well-known Husimi representation [23, 36, 37] of the laser pulse taken at the phase-space point $(\omega_n, t_m)$.

Suppose that the electric field is discrete and finite in the frequency domain, i.e. it is defined by $N$ complex values within the spectral range $\Omega = \omega_{\text{max}} - \omega_{\text{min}}$. Since it is discrete in frequency it is periodic in time with period $T = 2\pi N/\Omega$; thus the pulse (at least a unit cell of the pulse) can be described within the time–frequency rectangle determined by $T \otimes \Omega$. Intuitively, only a subset of the von Neumann lattice should be necessary to span this time–frequency rectangle, specifically the basis functions on the lattice sites that lie within (or on the boundary of) the rectangle. Since the criterion for completeness of the von Neumann basis is that each rectangular region of the lattice have area $2\pi$ [25], having $N$ von Neumann basis functions should provide a complete basis for a region of $2\pi N = \Omega T$, the area of the rectangle in time–frequency space. In order to treat time and frequency equivalently, we distribute these functions on a regular grid of $\sqrt{N} \times \sqrt{N}$ phase-space points covering the complete time span $T$ and frequency span $\Omega$ of the signal. The points $(\omega_n, t_m)$ corresponding to the centers of the basis functions are called von Neumann pixels. The width of the Gaussians in frequency are determined by the requirement that the standard deviation be $\Omega / \sqrt{2\pi N}$, corresponding to $\alpha = 2\pi N/(2\Omega^2) = T/(2\Omega)$.

The preceding paragraph implies that the information content in $N$ von Neumann basis functions should be equivalent to that in the original representation of $N$ points in frequency (or time). This implies that every signal defined in frequency domain and transformed to the von Neumann plane should be reconstructed exactly after back-transformation, a property which was qualitatively confirmed by the examples in [23]. However, small distortions of the reconstructed signal were noticeable in that work, the amplitude of the distortions depending on the spectral intensity of the input signal. In this paper, we analyze the origin of these distortions and then discuss several possible solutions to remove the distortions.
Figure 1. Test signal to examine the quality of the numerical implementation of the von Neumann transformation. (a) Complex Husimi representation in amplitude and phase. (b) Von Neumann representation in amplitude and phase. (c) Spectral amplitude of the original (red line) and the reconstructed (blue dotted line) signal. (d) Temporal amplitude of the original (red line) and the reconstructed (blue dotted line) signal.

3. Numerical implementation of the von Neumann representation and effects of truncation

The numerical implementation of the von Neumann transformation was performed by replacing the overlap integral in (4) with a sum over all $N$ points of the input signal, and by calculating the overlap matrix defined in (5) analytically and inverting it numerically.

It is clear that a necessary condition for a perfect reconstruction is that the truncated von Neumann lattice span the area in the time–frequency plane spanned by the signal. However, even if this condition is met we have found that most signals defined in the frequency domain lead to slightly distorted reconstructions. This is due to the fact that the complete von Neumann representation of these signals extends further than the finite von Neumann plane used for the present calculations. The most extreme error is where the input signal is a bandwidth-limited Gaussian laser pulse with the width of a von Neumann basis function, located between two von Neumann pixels in both time and frequency (red lines in figures 1(c) and (d)). The complex Husimi plot (figure 1(a)) is affected by this shift of half a pixel only in a trivial way: the amplitude in the complex Husimi representation is now spread over the nearest-neighbor pixels but pixels remote from the main peak have insignificant amplitude as would be expected given their insignificant overlap with the laser pulse. In the von Neumann representation, however, basis functions quite far away from the main peak contribute with alternating sign in adjacent pixels (figure 1(b)). This can be understood intuitively by imagining the construction...
of a Gaussian from other Gaussians whose centers are equidistantly distributed and do not coincide with the original one. Here, every positive contributing function has to be balanced by a negative contributing one further away and vice versa resulting in a large spread of amplitude in the von Neumann plane. This effect is caused by the nonorthogonality of the basis set used. The spreading of the amplitude over the complete von Neumann plane, although mathematically justified, complicates the intuitive interpretation and requires care to obtain an accurate reconstruction of the function.

Consider figure 1(c). When the joint time–frequency representation is transformed back to the frequency domain, distortions in the signal become visible (blue dotted line in figure 1(c)). The same is true for the temporal electric field (blue dotted line in figure 1(d)), which shows a train of small additional pulses appearing before and after the real pulse. Further iteration of the transformations does not alter the result, i.e. the distorted signal as well as the von Neumann representation stay conserved after repeated forward–backward transformations. This allows us to conclude that information is lost during the first transformation from the frequency domain to the finite von Neumann plane, which contradicts the claim that the von Neumann transformation is a one-to-one mapping. Since most of the points in the complex Husimi representation, which is the overlap integral between the signal and the von Neumann basis functions (compare (6)), have vanishing amplitude it appears that no relevant information has been discarded in the calculation of the overlap. Therefore, the information loss must be attributed to the appearance of the inverse overlap matrix $S_{(n,m),(i,j)}^{-1}$ in the calculation of the coefficients in the von Neumann representation (compare (4)) redistributing considerable information to von Neumann pixels outside the phase-space region one would have predicted based only on the Husimi overlaps.

4. Modified transformations

We developed three different approaches to solve this problem. They have different advantages and disadvantages with respect to future application in quantum control. We present a fourth, related approach, that does not help us solve the numerical problem but appears to be of interpretive value.

4.1. Use of a larger von Neumann plane

Since the problem of convergence seems to derive from the contribution of basis functions outside the finite joint time–frequency region, the simplest solution is to use a larger region for the calculation of the von Neumann representation.

The procedure is sketched in figure 2: starting from a discrete signal sampled at $N$ points in the frequency domain, the complex Husimi distribution is calculated on a $\sqrt{N} \times \sqrt{N}$ grid as described above using (6). Subsequently, $z$ additional lines of zeros are added in each direction resulting in a $(\sqrt{N} + 2z) \times (\sqrt{N} + 2z)$ grid for the complex Husimi representation. The overlap matrix $S_{(n,m),(i,j)}$ and its inverse as well as the von Neumann coefficients are then determined on this extended grid, allowing us to conserve information which under normal conditions would be cut away because it is contained in von Neumann pixels outside the original intervals $[\omega_{\text{min}}, \omega_{\text{max}}]$ and $[-T/2, T/2]$ of the joint time–frequency region of interest. For the reconstruction of the electric field the sum in (3) is carried out over all $(\sqrt{N} + 2z)^2$ basis functions. Since the expansion of the von Neumann plane is achieved by zero padding, i.e. adding lines of zeros, no additional information is added and the bijectivity of the von Neumann
transformation is conserved. The \( N \) points of the original signal are mapped one-to-one onto the \( N \) nonzero points of the complex Husimi representation. The reconstruction quality is improved the more additional lines of zeros are added. However, the improvement per additional line decreases while the efforts to calculate the inverse of the overlap matrix increase. In figure 3 the effect of zero padding is demonstrated using the test signal from figure 1. The black squares in figures 3(a) and (b) indicate the original size of the von Neumann grid for a signal sampling number of \( N = 121 \). Five additional lines of zeros have been added as a compromise between effort and benefit. The von Neumann amplitude is extending significantly into the additional phase-space region. As can be seen in figures 3(c) and (d), a much better reconstruction quality than in figures 1(c) and (d) is achieved. Since the spread of the signal in the von Neumann plane does not depend on the grid size, the number of required additional lines is essentially independent of the actual signal sampling number \( N \).

4.2. Introduction of periodic boundary conditions

The large computational effort involved in the previous approach can be avoided by implementing a quasi-infinite von Neumann plane using periodic boundary conditions.

The basis functions are altered to satisfy the periodicity condition \( \tilde{\alpha}_{\omega t m} (\omega + \Omega) = \tilde{\alpha}_{\omega t m} (\omega) \) in the frequency direction, i.e. the values of the basis functions leaking out on one side of the considered frequency region are reintroduced on the other side. Note, that the same periodicity in the time direction is already implicitly included due to the discrete sampling in the frequency domain\(^4\). This same periodicity will then enter into \( S \) and \( S^{-1} \) and ultimately into \( \tilde{Q} \). This enforced periodicity, together with calculating the overlap matrix \( S_{(m,m),(i,j)} \) numerically.

\(^4\) The periodicity in time is not clearly visible in figures 1 and 3 because the analytical overlap matrix used for those calculations neither had periodicity in time nor reflected the finite frequency range.
therefore allows the computation of the von Neumann representation to be strictly limited to the $N$ points of the joint time–frequency region of interest $\Omega \times T$ without any loss of information.

In figure 4, the same signal as in figures 1 and 3 is defined in the frequency domain (red lines in figures 4(c) and (d)), transformed to the von Neumann representation (figures 4(a) and (b)) and subsequently reconstructed (blue dotted line in figures 4(c) and (d)). In regions of significant amplitude, the agreement with the original signal is virtually exact, as can be seen in the perfect correspondence of the red lines and blue dotted lines in figures 4(c) and (d). The periodicity in time is now evident in the von Neumann amplitude plot, i.e. the part of the main peak that would be cut off at the edge of negative times is now visible at the edge of positive times. In frequency, the periodicity is less pronounced due to the larger distance of the main peak to the frequency edge.

4.3. Increasing the density of basis functions

Both aforementioned procedures require the inverse of the overlap matrix $S_{(n,m),(i,j)}$ in order to ensure a one-to-one mapping between frequency and joint time–frequency domain. This results in couplings not only between nearest-neighbor basis functions but even between those whose intensity overlap is negligible. Thus the interpretation of the corresponding phase-space representation is complicated. Furthermore, the periodicity may introduce even more of these counterintuitive couplings.

Since the complex Husimi representation (compare figure 2) introduced in (6) does not require the overlap matrix $S_{(n,m),(i,j)}$, it can be interpreted in a straightforward manner as
phase-space probability distribution. Hence for certain applications that do not necessitate the bijectivity of the transformation it might be desirable to use the complex Husimi directly rather than the von Neumann representation. In the one-to-one mapping, the resolution is identical to that of the von Neumann distribution. Reducing the phase-space volume per basis function from $2\pi$ to a lower value, i.e. using an overcomplete basis set, results in a more detailed phase-space picture at the cost of losing the bijectivity of the transformation. For such types of basis sets the overlap matrix becomes singular and can no longer be used to compensate for the nonorthogonality of the basis functions. However, if the density of basis functions is sufficiently high, the original signal can be obtained from the phase-space representation directly, without requiring the overlap matrix $S_{(n,m),(i,j)}$. Therefore the complex Husimi representation can be regarded as an intermediary between the discrete case, where an overlap matrix is required, and the continuous case where no overlap matrix is needed.

This procedure is demonstrated in figure 5, where the density of basis functions has been increased fourfold with respect to the representations from figures 3 and 4, resulting in a more detailed phase-space picture (figures 5(a) and (b)). In fact, as mentioned before, the absolute value squared of this representation is the well-known Husimi distribution function of the optical field and can be regarded as the detection probability distribution in time and frequency. A perfect signal reconstruction is achieved by this increased basis function density, as can be seen from the agreement between the original and the reconstructed signal (red and blue dotted lines in figures 5(c) and (d), respectively). The increased level of detail comes at the cost of having

**Figure 4.** Test signal from figure 1 using periodic boundary conditions: Von Neumann representation in (a) amplitude and (b) phase. (c) Spectral amplitude of the original (red line) and the reconstructed (blue dotted line) signal. (d) Spectral phase of the original (red line) and the reconstructed (blue dotted line) signal.

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more phase-space points, thus losing the bijectivity of the transformation. Therefore in contrast to the methods described in sections 4.1 and 4.2, this method is not suited for the definition of pulses in phase-space. However, due to its higher resolution it is well adapted for representing shaped laser pulses and analyzing them with respect to their ‘mechanistic building blocks’.

4.4. Use of the complex Husimi on a von Neumann grid

It is noteworthy that the complex Husimi is calculated as an intermediate step toward the von Neumann representation as mentioned in section 3 and illustrated in figure 2. In contrast to the preceding section 4.3, it is here used on a von Neumann grid. Although the spacing in phase space is much coarser, it still allows an interpretation as a measurement probability distribution. We present this approach not as a possible solution to numerical problems introduced by truncation of the phase-space basis, but because we believe it has promise as an interpretive tool.

The method is illustrated in figure 1(a) where the complex Husimi representation of a Gaussian test signal is shown as amplitude and phase. In contrast to the von Neumann representation (figure 1(b)), where considerable amplitude is distributed over the complete von Neumann grid, the amplitude of the complex Husimi representation is clearly localized in phase space and has a Gaussian-shaped distribution. As mentioned above, the complex Husimi representation on a von Neumann grid is only an intermediate step in the calculation of the
corresponding von Neumann representation. The achieved reconstruction quality consequently depends solely on how this intermediate data are processed, figures 3(c), (d) and 4(c), (d) representing two examples of such processing.

Since each individual pixel of the complex Husimi representation is the overlap integral between the signal and the corresponding Gaussian basis function, the pixel itself no longer corresponds to a bandwidth-limited laser pulse. This, together with using the methods introduced in sections 4.1 and 4.2, ensures the bijectivity of the mapping between the complex Husimi and the frequency domain representations. Since the inverse overlap matrix \( S^{-1}_{n,m}(i,j) \) is not involved in the calculation of the complex Husimi representation (compare (6)), it is therefore possible to define laser pulses directly using the physically meaningful measurement probability distribution avoiding the unintuitive couplings present in the von Neumann distribution.

5. Conclusions

The problem of the imperfect reconstruction of signals in the finite sized and discrete von Neumann transformation can be solved using several approaches. Depending on the actual application, the most suitable method can be selected. It is possible either (i) to extend the used von Neumann grid, (ii) to introduce explicit periodic boundary conditions or (iii) to increase the density of basis functions. In all cases, the reconstruction quality is excellent. In the present work, we have chosen a particular pulse shape for illustration. In the selected case, the reconstruction is especially difficult because the pulse center is located between different von Neumann pixels. But what about other pulse shapes? When using periodic boundary conditions (section 4.2) we found that any arbitrary pulse shape is perfectly reconstructed. No difference in accuracy could be found for different pulse shapes. The approaches of using a larger von Neumann plane (section 4.1) and of increasing the density of basis functions (section 4.3) are of a different nature. Here the achieved accuracy depends on the chosen size of the extended von Neumann plane or on the chosen basis function density, respectively. These parameters therefore have to be set manually to achieve a desired accuracy. When the extended von Neumann plane is too small or the increased basis function density too low, there still remains some (small) inaccuracy that depends on the chosen pulse shape. Qualitatively, the smaller the overlap between the defined pulse shape and the available basis functions, the higher is the remaining error. However, when the extended von Neumann plane size or the basis function density are chosen appropriately such that machine accuracy is reached, we could not find any differences in the reconstruction quality for different pulse shapes.

In the field of quantum control of chemical reactions with shaped femtosecond laser pulses, adaptive learning algorithms are often used to find the optimal pulse shape for a given control target [4, 10, 38]. As discussed in [24], this technique can be implemented using a joint time–frequency representation to obtain an intuitive understanding of the mechanism behind the optimal pulse shape. For that purpose it is important to retain a one-to-one mapping between the frequency domain, in which the pulse shaper is addressed, and the joint time–frequency representation, in which the laser pulses are parameterized, since the number of degrees of freedom of the search space is determined by the pulse-shaping apparatus [39]. Additionally, for an experimental implementation the computational effort should be kept as small as possible. The requirement of a one-to-one mapping is fulfilled either by using the approach of an extended von Neumann grid or periodic boundary conditions. Since the former involves a significantly
increased computational effort, the use of periodic boundary conditions is favored. On the other hand, the complex Husimi representation offers the best interpretation and can even be used together with periodic boundary conditions on a von Neumann grid where the phase-space volume per basis function is $2\pi$.

Another possible application of joint time–frequency distributions in quantum control is the use of ‘mechanistic building blocks’ for pulse construction where the individual blocks correspond to transform-limited Gaussian laser pulses centered around different points in time and frequency. This can be understood in terms of the frequently used Tannor–Kosloff–Rice scheme of quantum control [5, 6], in which multiple pulses transfer population in suitable Franck–Condon windows at times determined by the wave-packet dynamics. Therefore, the von Neumann representation, where the individual pixels correspond to such building blocks, is most likely to supply the right choice of parameterization. Since the computational effort involved in extending the von Neumann grid used is significantly higher than with periodic boundary conditions the latter solution is the favorable choice. If an interpretation of the shaped laser pulses as a measurement probability in the joint time–frequency domain is preferred, the complex Husimi representation on a von Neumann grid can be used together with periodic boundary conditions.

Although not offering a one-to-one mapping, the use of the complex Husimi with an increased density of basis functions is an appropriate representation to analyze shaped pulses in general. It is noteworthy that in contrast to the ordinary Husimi representation the complex counterpart still contains all the phase information [23].

In summary, we believe that the von Neumann representation and its modified versions introduced here can be very useful for improved coherent control experiments, especially, considering the enormous utility of the closely related Gabor transform in the field of signal processing. In particular, the method is expected to (i) provide mechanistic insight into the function of complicated shaped pulses, (ii) allow for automated scanning of multicolor multidimensional coherent spectroscopy, and (iii) lead to improved ‘intelligent’ evolutionary algorithms whose genes are mechanistic building blocks.

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