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Nonuniform sampling by quantiles

D. Levi Craft [a], Reilly E. Sonstrom [a], Virginia G. Rovnyak [b], David Rovnyak [a,*]

[a] Department of Chemistry, Bucknell University, Lewisburg, PA 17837, United States
[b] University of Virginia School of Nursing, Charlottesville, VA 22908, United States

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ABSTRACT

A flexible strategy for choosing samples nonuniformly from a Nyquist grid using the concept of statistical quantiles is presented for broad classes of NMR experimentation. Quantile-directed scheduling is intuitive and flexible for any weighting function, promotes reproducibility and seed independence, and is generalizable to multiple dimensions. In brief, weighting functions are divided into regions of equal probability, which define the samples to be acquired. Quantile scheduling therefore achieves close adherence to a probability distribution function, thereby minimizing gaps for any given degree of subsampling of the Nyquist grid. A characteristic of quantile scheduling is that one-dimensional, weighted NUS schedules are deterministic, however higher dimensional schedules are similar within a user-specified jittering parameter. To develop unweighted sampling, we investigated the minimum jitter needed to disrupt subharmonic tracts, and show that this criterion can be met in many cases by jittering within 25–50% of the subharmonic gap. For nD-NUS, three supplemental components to choosing samples by quantiles are proposed in this work: (i) forcing the corner samples to ensure sampling to specified maximum values in indirect evolution times, (ii) providing an option to triangular backfill sampling schedules to promote dense/uniform tracts at the beginning of signal evolution periods, and (iii) providing an option to force the edges of nD-NUS schedules to be identical to the 1D quantiles. Quantile-directed scheduling meets the diverse needs of current NUS experimentation, but can also be used for future NUS implementations such as off-grid NUS and more. A computer program implementing these principles (a.k.a. QSched) in 1D- and 2D-NUS is available under the general public license.

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1. Introduction

Nonuniform sampling in NMR is often regarded as the practice of acquiring data over indirect evolution periods that are not equally spaced in time. The benefits of NUS include reducing experimental time, improving resolution, improving spectral bandwidth, avoiding Fourier artifacts through the use of alternative reconstruction methods [1–3], and enabling the study of much larger proteins [4]. By weighting the density of samples to a decaying or other nonconstant signal envelope, the signal-to-noise ratio of the time domain data can be improved [2,5–11], is never worse than for time-equivalent uniform sampling, and may even be improved by factors up to as much as two fold in favorable cases in a given dimension [12,13]. The utility of NUS in direct evolution periods has also been developed to reduce power requirements and to facilitate homonuclear decoupled spectroscopy [14,15].

Many approaches to nonuniform sampling have been practiced in NMR for several decades and, as researchers strive to get the most benefits out of NUS, the strategies for choosing a nonequidistant series of samples, referred to as a sampling schedule, have come under increasing scrutiny. Specifically, spectral quality and reproducibility can be promoted by satisfying a number of criteria for the design of the sampling schedule. First, the understanding that any spectral estimation technique is faced with deconvolving the 'spectrum' of the sampling schedule, known as the point spread function (PSF) [2,3], from the desired experimental spectrum means that it is important to have randomness in the sampling schedule [2,16–18], which could be addressed by randomly jittering samples within distinct areas of the sampling space [17]. However, avoiding large gaps in distributing samples over the evolution period is also important, as large gaps can be viewed as local regions of extremely sparse sampling, which may impact spectral aliasing or present challenges in spectral reconstruction [17,19]. Minimizing or effectively eliminating folding within the Nyquist bandwidth is also required [20,21], and can be achieved by including some uniformly spaced tracts in the sampling schedule, by avoiding schedules that are too sparse, and by choosing NUS from an over-sampled Nyquist grid [2,22]. Weighting the sampling generally means that more data are acquired in certain regions of the

* Corresponding author.
E-mail address: drovnyak@bucknell.edu (D. Rovnyak).

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signal evolution, such as when the signal is strongest, promoting efficient use of spectrometer time as well as sensitivity gains [2,4–9,11–13,23–25]. Line shapes in spectra reconstructed from NUS data are often indistinguishable from their appearances in spectra obtained via uniform sampling and the FFT; but when NUS is sparse or strongly weighted, then line shapes may be perturbed to varying degrees [26]. Objective statistically derived metrics for evaluating the performance of NUS schedules have also been recently devised [27].

The simplest approach to generating NUS schedules, that of choosing a series of random numbers according to a probability density, yields highly variable, seed-dependent sampling schedules, often exhibiting large gaps and poor point spread function characteristics, that can degrade the quality of spectral reconstructions [26,28–31]. While a number of recent methods exist for generating more robust NUS schedules [1,17,19,26,28,30–33], we perceived a need to propose an approach to NUS scheduling based on the use of statistical quantiles, which define regions of equal probability in a sampling density, that will (i) bring together known criteria for optimizing sampling schedules; (ii) offer features that we perceive are not available in current packages; (iii) be an intuitive and theoretically grounded method that we hope will promote adoption of NUS by a broader base of scientists; and (iv) be a flexible foundation for future NUS implementations, such as choosing samples with greater time precision than the Nyquist grid. Importantly, some of the most recently devised sampling strategies have been evolving towards the goal of choosing samples determined by regions of equal probability [17,26,30–32], suggesting that multiple independent efforts are converging along different paths towards the quantile concept. The present approach attempts to consolidate these efforts by explicitly starting with quantiles of the probability density to direct the choice of nonuniform samples. This work shows that quantile-directed NUS is flexible for weighted and unweighted sampling, and represents a general strategy for n-dimensional NUS. A software program (a.k.a. QSched) accompanies this manuscript; information on obtaining the program is given in the Supplemental Information. Partial component sampling [34], beat-matched sampling [11], and oversampled/off-grid applications are not currently implemented but will be considered in future releases.

Addressing reproducibility in NUS is gaining increased attention, but is facing definitional differences. One approach is that generating schedules for nonuniform sampling that do not depend significantly on a random seed can be viewed as a form of reproducibility [28,30]. In such a case, reproducibility is taken to mean that if two sampling schedules were generated with identical parameters, but still differ from each other, then the resulting spectral estimates from each of the two schedules will have the same spectral information within reasonable uncertainty estimates. This approach focuses on the preservation of spectral information as the condition for reproducibility, and also implies that the spectral information should be accurate. However it may be more useful to classify this approach as 'low variance' sampling (private communication: Prof. A. Schuyler). Reproducibility can have a distinct meaning, that the sampling schedule can be generated exactly by other investigators, which may be achieved through deterministic scheduling or by managing the random seeds employed. This approach decouples reproducibility from criteria related to the spectral estimates, and instead seeks to ensure that schedules can be generated exactly, even if the schedule itself is lost. We do not undertake here to clarify the direction of this important discussion, but will point out how quantile-directed scheduling relates to principles that can be found in both approaches to reproducibility.

Results obtained here show that quantile-directed sampling promotes seed-independence: deterministic schedules are generated for weighted NUS in one dimension (e.g. 2D NMR), while for multidimensional NUS such as 2D-NUS (e.g. 3D NMR), nondeterministic schedules are generated that have a high degree of similarity due to the use of a centered, constrained jitter, which shares some features with random sampling algorithms developed by Kazimierczuk [17] and Mobli [30]. In other words, quantile-directed nD-NUS schedules generated with the same parameters will not be identical, but will be forced to be similar within the constrained jitter. A special case of quantile-directed sampling arises for unweighted NUS in one dimension, where schedules will not be deterministic due to the need for a constrained jitter to disrupt subharmonic sequences.

2. Theory and background

A specific evolution time that is recorded in an NMR experiment is termed a sample, and a list of such discrete evolution times in an NMR experiment is a sampling schedule. The sampling schedule may be chosen uniformly using equidistant time intervals, or nonuniformly using time intervals that are not all equal. Sampling may be on-grid or off-grid, referring to whether the samples are a subset of the original uniform time series (on-grid, a.k.a. the Nyquist grid) or if they are chosen at times with precision that is limited primarily by hardware timing specifications [20]. This work considers on-grid NUS, but was designed with future applications to off-grid NUS in mind.

In this work, a sample will consist of the real and imaginary parts in each indirect dimension. Specifically, for one indirect evolution period, a 'sample' refers to both the real and imaginary FIDs acquired for each evolution time. Similarly, for two indirect periods, a 'sample' refers to all four FIDs (RR, RI, IR, II) acquired at a given evolution time. The practice of selecting only the real or imaginary parts at a given evolution time, known as partial component sampling (PCS), can achieve more coverage of evolution times and improved randomization of the sampling schedule [34], but is beyond the scope of the current work.

A one-dimensional probability density function (PDF) is a function of time, $\rho(t)$, which, for any interval, determines the probability that a result will lie within the interval. The PDF is often presented on a scale so that the area under the PDF is 1, and the probability that a result will lie within an interval is the area under the PDF over the interval. In nonuniform sampling, a PDF describes the probability of choosing a sample in an NMR evolution period. If there is more than one independent evolution time, a PDF may be multidimensional, e.g. $\rho(t_1, t_2, \ldots)$.

Two cases for the PDF will be considered. A PDF may be constant, meaning that the probability of choosing a sample is the same throughout the evolution period, termed unweighted sampling. Or a PDF may be nonconstant, meaning that the probability of sampling the evolution period is greater at some times than others, called weighted sampling.

For NMR spectroscopy, selecting samples according to a PDF in one dimension (e.g. NUS in one dimension of nD-NMR) should be distinguished from selecting samples from a multidimensional PDF (e.g. NUS in two or more dimensions of nD-NMR). The strategy of quantile-determined scheduling affords a great deal of flexibility to design NUS schedules for a wide variety of experimentation. Three cases, summarized here, demonstrate the broad scope of quantile-directed sampling, and will be developed in the Results and Discussion.

I. *Weighted 1D-NUS (*e.g. *2D-NMR).* Deterministic schedules result directly from the use of quantiles.
II. *Unweighted 1D-NUS*. Quantiles of a uniform probability distribution are subjected to a user-specified constrained jitter to improve randomness and to enforce the bandwidth given

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