RB-SFA: High Harmonic Generation in the Strong Field Approximation via *Mathematica*

Usage and Examples

Loading the package

You can use this software

- within the RB-SFA notebook itself by simply running the initialization cells of that notebook, or
- from an external notebook by loading it as a package.

In the latter case, place a copy of the package file RB-SFA.m on the same directory as your notebook and run the loading command

```
Needs["RBSFA`", FileNameJoin[{NotebookDirectory[], "RB-SFA.m "}]]
```

You can also call the package from another directory by suitably modifying the directory call. If you plan on using this package in the long term you can use the File > Install prompt, in which case the package is simply loaded as Needs[“RBSFA`”], though this is not particularly recommended.

Simple usage

For basic usage, simply call the main numerical integrator, makeDipoleList, with the vector potential you want to use, and provide any parameters you wish to specify using the FieldParameters option.

```
AbsoluteTiming[
  simpleDipole = makeDipoleList[VectorPotential→Function[t, {F/ω Sin[ω t], 0, 0}],
      FieldParameters →{F→0.05, ω→0.057}];
]
```

```
{1.99374, Null}
```

Calling the function with insufficient parameters will produce error messages:

```
makeDipoleList[VectorPotential→Function[t, {F/ω Sin[ω t], 0, 0}]]
```

```
makeDipoleList::pot :
   The vector potential A provided as VectorPotential→Function[t, {F Sin[ω t]/ω, 0, 0}] is incorrect or is missing
      FieldParameters . Its usage as A[3.482993423326028`] returns
      {F Sin[3.48299 ω]/ω, 0, 0} and should return a list of numbers .
```

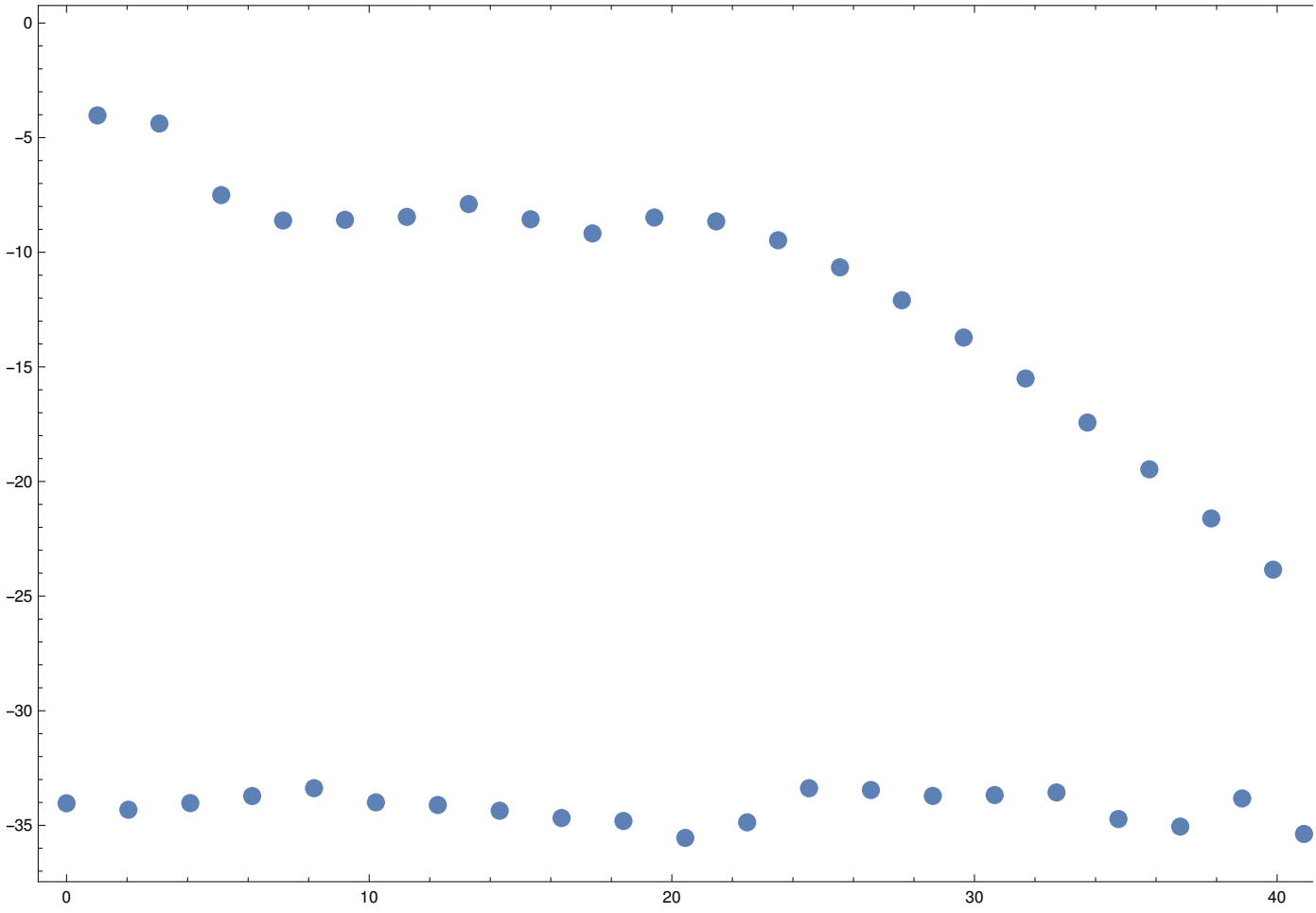
```
$Aborted
```

The symbol ω is taken to be the carrier frequency, and is set by default to $\omega = 0.057$ atomic units, corresponding to a wavelength of 800 nm. If the carrier frequency is changed, this must be specified on **both** the field parameters and the explicit option for the integrator, as

```
makeDipoleList [VectorPotential→ Function[t, {F/ω Sin[ωt], 0, 0}],
  FieldParameters →{F→0.05, ω→0.0456}, CarrierFrequency→0.0456]
```

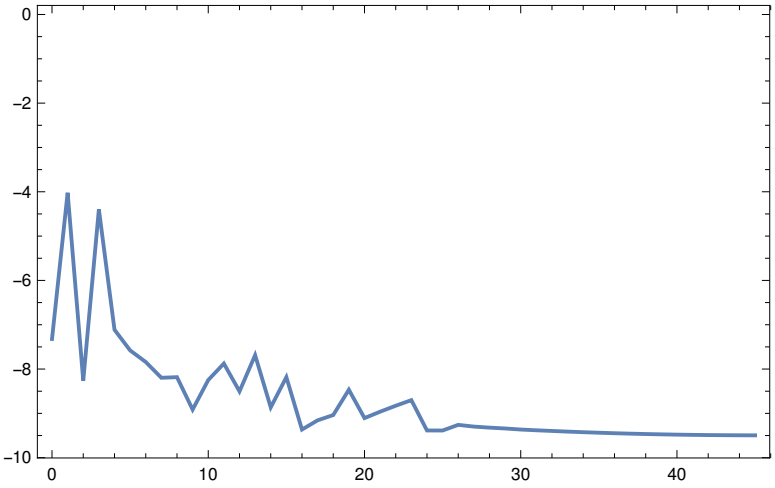
To see the spectrum, use the getSpectrum and the spectrumPlotter commands, such as

```
spectrumPlotter [getSpectrum [Most[simpleDipole]], Joined→False]
```



Note here the use of Most on the dipole when a monochromatic field is indicated. This ensures that the signal is actually periodic (i.e. it eliminates repetition between the initial and final points, which are separated by exactly one period). If this is not done, the spectrum is much noisier:

```
spectrumPlotter [getSpectrum [simpleDipole], ImageSize → 400]
```



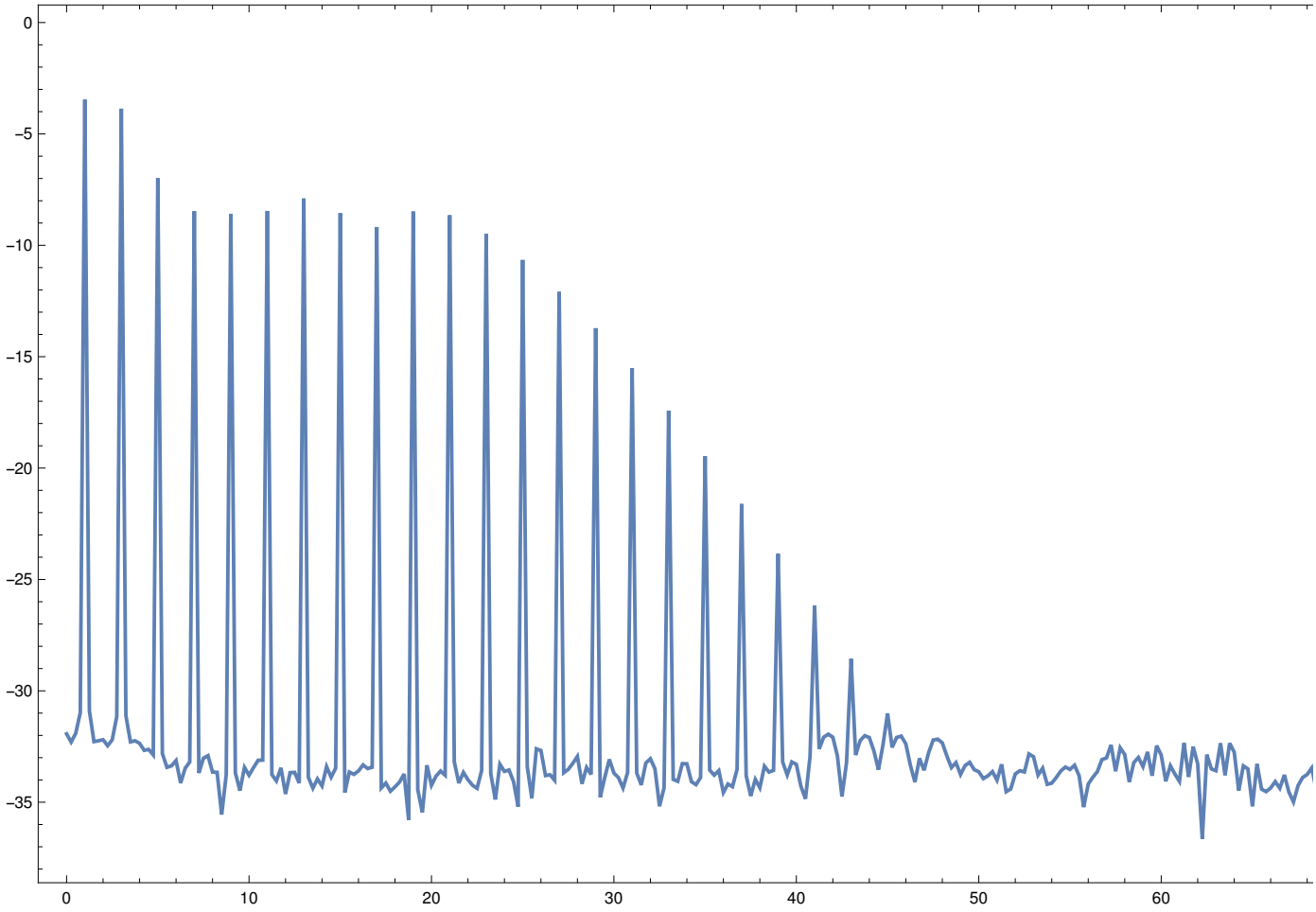
The default options are built for a periodic pulse for which simple functions of the vector potential can be integrated analytically, and for which only a single period of integration is necessary. More periods can be specified using the TotalCycles option. Similarly, the PointsPerCycle option controls the number of points per period.

```
AbsoluteTiming [
  biggerDipole = makeDipoleList [VectorPotential → Function[t, {F/ω Sin[ω t], 0, 0}],
      FieldParameters → {F → 0.05, ω → 0.057}, TotalCycles → 4, PointsPerCycle → 150];
]
```

```
{22.4014, Null}
```

To get a correct spectrum plot, give these settings to the spectrum plotter.

```
spectrumPlotter [getSpectrum [Most[biggerDipole]], TotalCycles→4, PointsPerCycle→150]
```



You can specify a Target chemical species using the option

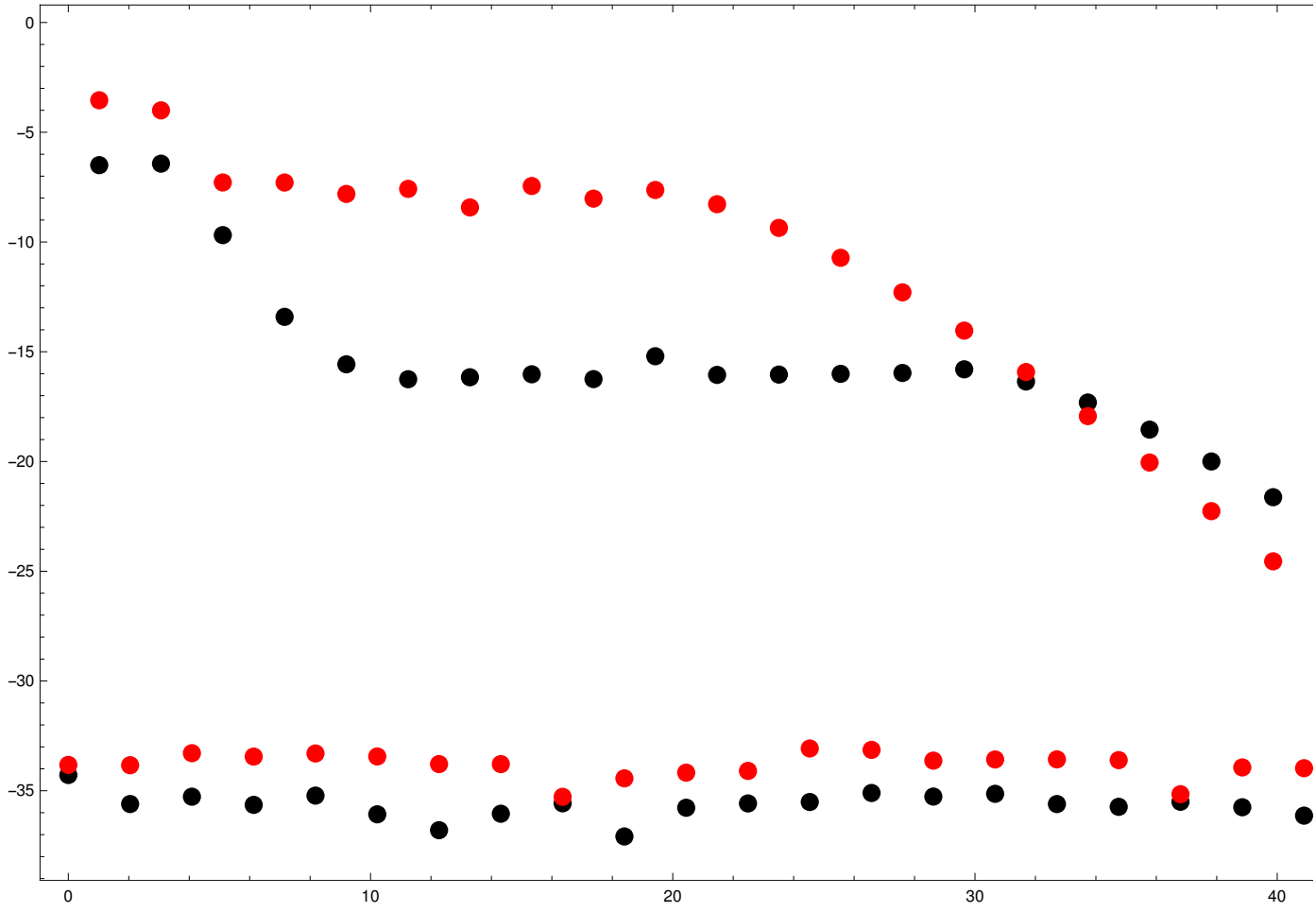
```
?Target
```

Target is an option for makeDipoleList which specifies chemical species producing the HHG emission, pulling the ionization potential from the Wolfram ElementData curated data set.

i.e. using the syntax

```
heliumDipole = makeDipoleList [VectorPotential→Function[t, {F/ω Sin[ω t], 0, 0}],
      FieldParameters →{F→0.05, ω→0.057}, Target→"Helium "];
xenonDipole= makeDipoleList [VectorPotential→Function[t, {F/ω Sin[ω t], 0, 0}],
      FieldParameters →{F→0.05, ω→0.057}, Target→"Xenon"];
```

```
Show[{
    spectrumPlotter [getSpectrum [Most[heliumDipole ]], Joined→ False, PlotStyle → Black],
    spectrumPlotter [getSpectrum [Most[xenonDipole]], Joined→ False, PlotStyle → Red]
  }]
```



An ionization potential can also be specified directly:

```
?IonizationPotential
```

IonizationPotential is an option for makeDipoleList which specifies the ionization potential I_p of the target.

To see the available options for this function (and others), use

```
Options[makeDipoleList ]
```

```
{PointsPerCycle→ 90, TotalCycles → 1, CarrierFrequency→ 0.057,
  VectorPotential→ Automatic , FieldParameters → {}, VectorPotentialGradient→ None,
  Preintegrals→ Analytic, ReportingFunction→ Identity, Gate → SineSquaredGate[1/2],
  nGate → 3/2, ϵCorrection→ 0.1, IonizationPotential→ 0.5, Target → Automatic ,
  DipoleTransitionMatrixElement→ hydrogenicDTME, PointNumberCorrection → 0, Verbose → 0}
```

All options have suitable information messages.

```
?VectorPotential
```

VectorPotential is an option for makeDipole list which specifies the
field's vector potential. Usage should be VectorPotential→A, where A[t]//.pars must yield
a list of numbers for numeric t and parameters indicated by FieldParameters→pars.

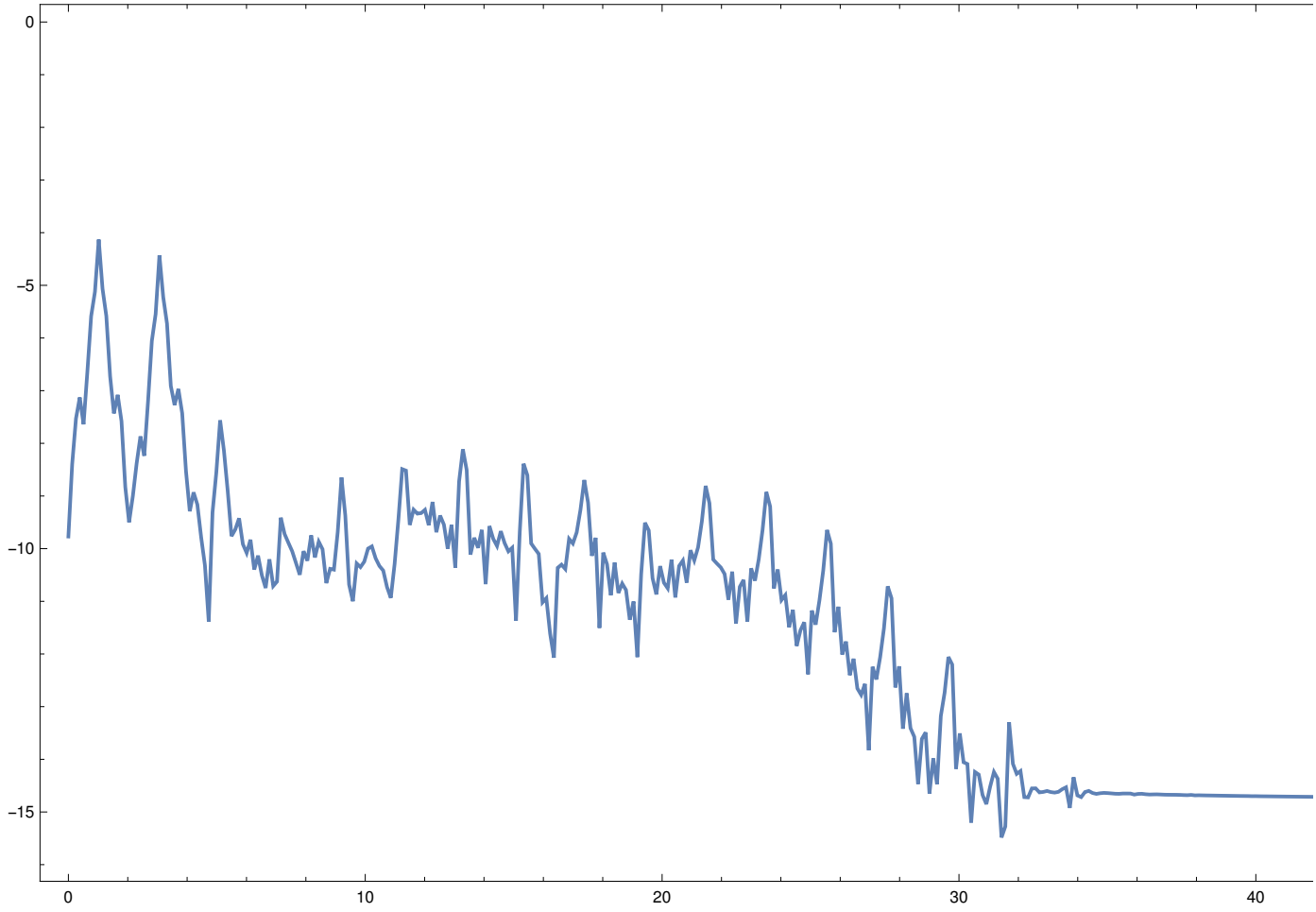
Using numerical integration

To simulate a pulse with an envelope, it can be convenient to perform the preintegrals numerically, using the option Preintegrals→"Numeric". These cases are generally slower but mainly because they require many more periods of integration.

```
AbsoluteTiming [
  numericallyIntegratedDipole =
      makeDipoleList [VectorPotential→ Function[t, {F/ω envelope[t] Sin[ω t], 0, 0}],
        FieldParameters →{ω→0.057, F→0.055, envelope→cosPowerFlatTop[0.057, 8, 16]},
        TotalCycles→8, Preintegrals→"Numeric "];
]
```

```
{16.0119, Null}
```

```
spectrumPlotter [getSpectrum [numericallyIntegratedDipole ]]
```



When using flat top pulses, and other waveforms that depend on Piecewise functions, it is possible that the function

will return errors caused by an Indeterminate derivative being evaluated at the corners of the envelope.

```
AbsoluteTiming [
  flatTopPulseDipole=
      makeDipoleList [VectorPotential→ Function[t, {F/ω envelope[t] Sin[ω t], 0, 0}],
        FieldParameters →{ω→0.057, F→0.055, envelope→flatTopEnvelope[0.057, 8, 2]},
        TotalCycles→8, Preintegrals→"Numeric "];
]
```

```
{17.7842, Null}
```

In these cases, use a numeric test to diagnose what's happened

```
Tally[flatTopPulseDipole/. _?NumberQ →✓]
```

```
{{{✓, ✓, ✓}, 721}}
```

and if the function is returning non-numeric values, it can help to fiddle with the PointNumberCorrection option.

```
?PointNumberCorrection
```

PointNumberCorrection is an option for makeDipole list which specifies an extra number of points to be integrated over, which is useful to prevent Indeterminate errors when a Piecewise envelope is being differentiated at the boundaries.

Parallelized environments

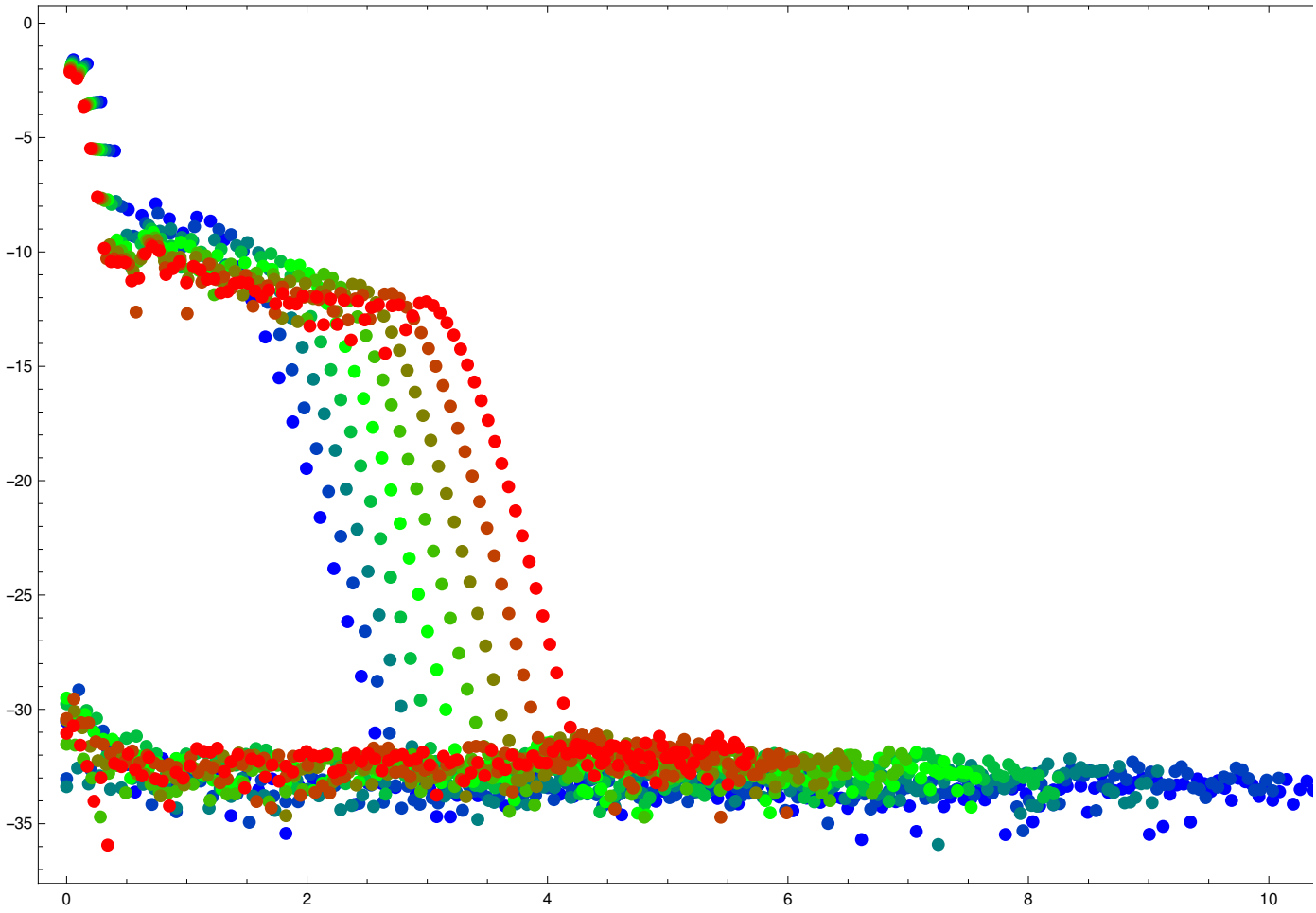
The numerical integration can be parallelized over via the use of ParallelTable and similar commands. Care must be taken to ensure that all parallel kernels have the package definitions available (using DistributeDefinitions, ParallelNeeds, or similar constructs). If a variable or function is used to store the results, this must be synchronized using SetSharedFunction or SetSharedVariable, as usual.

```
DistributeDefinitions["RBSFA`"];
SetSharedFunction[wavelengthScanDipole];

ParallelTable[
  Print[AbsoluteTiming [
      wavelengthScanDipole[λ] =
          makeDipoleList [VectorPotential→ Function[t, {F/ω Sin[ω t], 0, 0}], FieldParameters →
              {F→0.05, ω→45.6/λ}, CarrierFrequency→45.6/λ, PointsPerCycle→400];
    ]]
  , {λ, 800, 1600, 100}]
```

```
{65.3966, Null}
{67.5219, Null}
{67.9824, Null}
{68.8184, Null}
{70.7804, Null}
{70.9155, Null}
{71.6026, Null}
{43.4543, Null}
{41.8719, Null}
{Null, Null, Null, Null, Null, Null, Null, Null, Null}
```

```
Show[Table[
    spectrumPlotter [getSpectrum [Most[wavelengthScanDipole[λ]]],
      PlotStyle→Blend[{Blue, Green, Red}, λ/800-1], CarrierFrequency→45.6/λ,
      Joined→False, FrequencyAxis→"Frequency", PointsPerCycle→400]
    , {λ, 800, 1600, 100}]]
```



Writing output to file

For very large calculations (many integration points per cycle, in particular), the limiting factor is available memory. In these situations, it can help to write the data directly to a file on disk. This is slower (by a factor of about 2) but it

has a roughly constant RAM footprint, so it enables calculations of a bigger size than would be possible otherwise. (Of course, this can also be done from non-parallelized calls!) This is done via the ReportingFunction option:

```
?ReportingFunction
```

```
ReportingFunction is an option for makeDipole list which specifies a function
    used to report the results, either internally (by the default, Identity) or to an external file.
```

In essence, the integration loop consists of a Table construct, which goes over the time *t* at which the integral is performed, and an inner integration construct. Setting an option ReportingFunction→f interposes the function f between these two steps, as

```
Table[ f[ integrator[t]  ]  , {t, tInitial, tFinal}]
```

The default is f=Identity, which returns its input untouched, but it can also be replaced by a Write construct that can shunt its input to the hard disk without telling the kernel what it is, so it is not kept in memory.

```
Quit
```

```
DistributeDefinitions["RBSFA`"];
directory = NotebookDirectory[];
filename [F_] :=
    FileNameJoin [{directory, "Field scan data at F=" <> ToString[F] <> ".txt"}];

ParallelTable[
  Print[AbsoluteTiming [
      makeDipoleList [VectorPotential → Function[t, {F/ω Sin[ω t], 0, 0}],
          FieldParameters → {ω → 0.057}, CarrierFrequency → 0.057, PointsPerCycle → 400,
          ReportingFunction → Function[Write[filename [F], #]]
        ];
    ]]
  , {F, 0.05, 0.2, 0.025}]
```

```
{66.2765, Null}
{68.2327, Null}
{68.4573, Null}
{69.0505, Null}
{69.6457, Null}
{69.8211, Null}
{70.4778, Null}
{Null, Null, Null, Null, Null, Null, Null}
```

The data in the files can then be pulled in quite simply using e.g.

```
Do[ intensityScanDipole[F] = ReadList[filename [F]], {F, 0.05, 0.2, 0.025}]
```

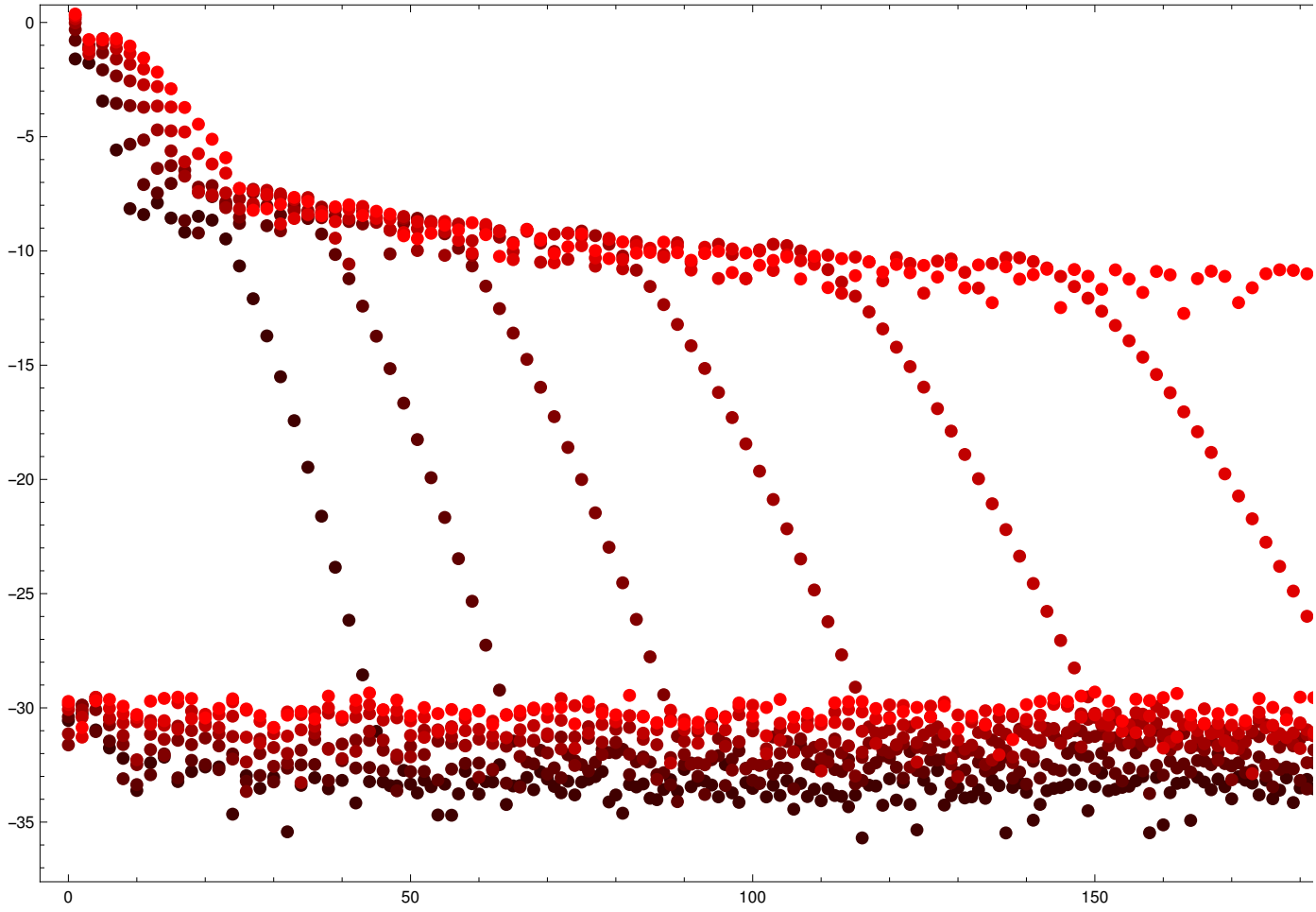
This tends to litter the directories by creating lots of files for different parameters, so it is usually cleaner to Save them into a single file, e.g. using

```
Save[FileNameJoin[{NotebookDirectory[], "Field scan collected data.txt"}],
  intensityScanDipole]
```

which in turn can then be pulled in using

```
<< (FileNameJoin[{NotebookDirectory[], "Field scan collected data.txt"}]);
```

```
Show[Table[
    spectrumPlotter[getSpectrum[Most[intensityScanDipole[F]]], CarrierFrequency→0.057,
      Joined→False, PointsPerCycle→400, PlotStyle→Blend[{Black, Red}, F/0.2]]
    , {F, 0.05, 0.2, 0.025}]]
```



As written, though, this has the disadvantage that each subkernel must access the hard drive for *every* timestep of the computation, which obviously responsible for (at least most of) the slowdown. A middle ground is also possible by choosing an appropriate ReportingFunction: a function which will cache a specific number *k* of results on RAM, and then write them to file all in one go. This is on the development to do (wish) list, and will hopefully be implemented soon - if time allows.

Nondipole contributions

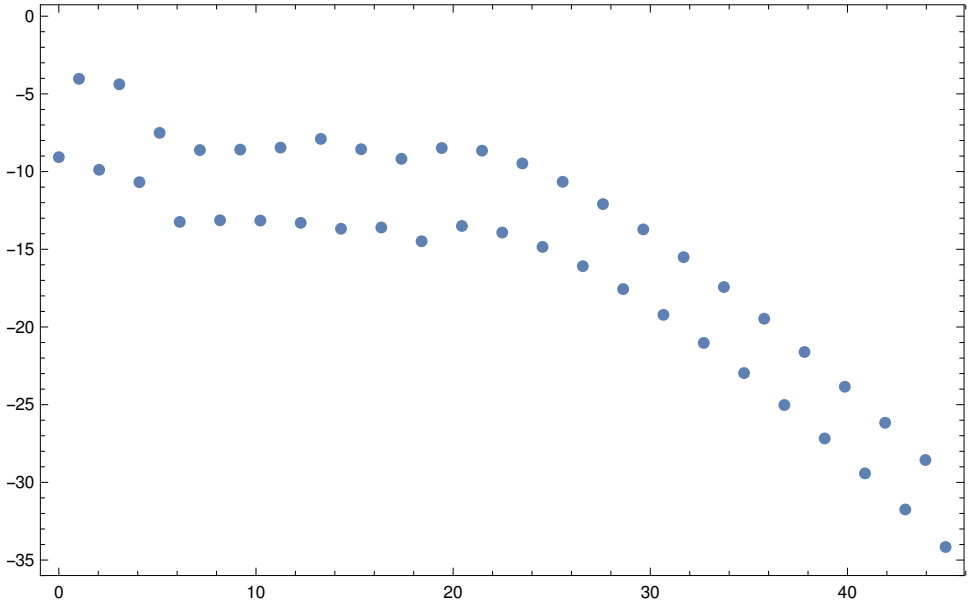
Nondipole contributions can be specified by setting a nonzero vector potential gradient:

```
?VectorPotentialGradient
```

"VectorPotentialGradient is an option for makeDipole list which specifies the gradient of
the field's vector potential. Usage should be VectorPotentialGradient→GA, where GA[t]//.pars must
yield a square matrix of the same dimension as the vector potential for numeric t and parameters
indicated by FieldParameters→pars. The indices must be such that GA[t][[i,j]] returns $\partial_i A_j[t]$."

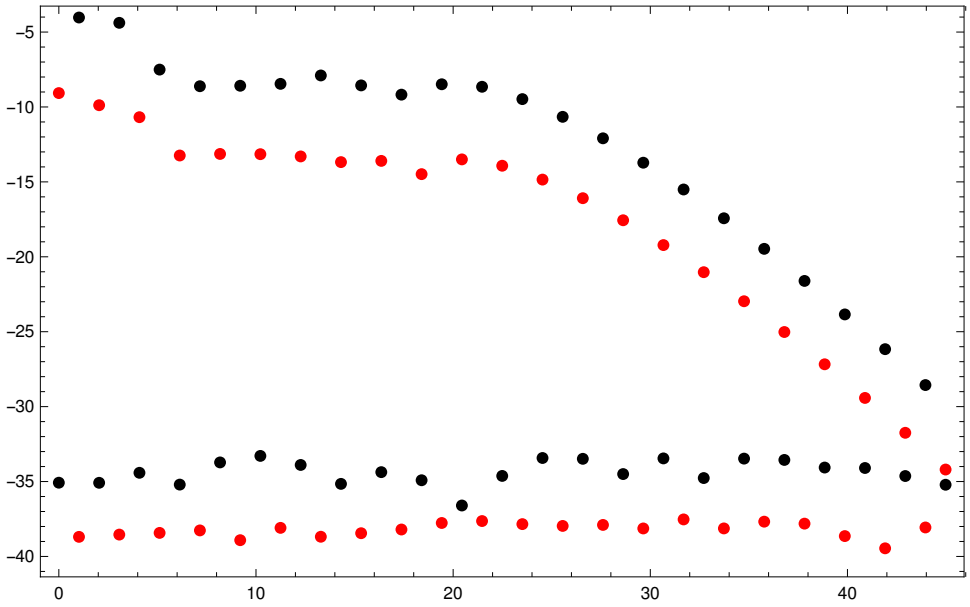
If, for example, the travelling-wave form of the vector potential is of the form $\boldsymbol{A}(\boldsymbol{r}, t) = \frac{F}{\omega}\hat{\boldsymbol{x}}\cos(k z - \omega t)$, then at the origin the vector potential is $\boldsymbol{A}(\boldsymbol{0}, t) = \frac{F}{\omega}\hat{\boldsymbol{x}}\cos(\omega t)$ and it has a single nonzero entry in its gradient matrix $\nabla \boldsymbol{A}$, i.e. $\partial_z A_x = -\frac{kF}{\omega}\sin(\omega t)$. This is entered into the VectorPotentialGradient option as

```
nonDipoleContributions = makeDipoleList [
    VectorPotential→ Function[t, {F/ω Cos[ω t], 0, 0}],
    VectorPotentialGradient→ Function[t, {{0, 0, 0}, {0, 0, 0}, {-k F/ω Sin[ω t], 0, 0}}],
    FieldParameters → {F → 0.05, ω → 0.057, k → ω / c, c → 137}
  ];
spectrumPlotter [getSpectrum [Most[nonDipoleContributions$]], Joined→ False, ImageSize → 500]
```



At low wavelengths, the first obvious effect is the appearance of even harmonics. This is the expected behaviour, with the harmonics along the laser propagation direction. (Informally, the magnetic pushing on the wavepacket acts on the propagation direction on both halves of each laser period. This off-axis recollision causes the dipole to oscillate in the propagation direction with an even symmetry. More formally, the dynamical symmetries of the problem permit even (but not odd) harmonics along this direction.) This is indeed what is observed:

```
Show[{
    spectrumPlotter [getSpectrum [nonDipoleContributions[1 ;; -2, {1, 2}]],
      Joined→ False, PlotStyle→ Black],
    spectrumPlotter [getSpectrum [nonDipoleContributions[1 ;; -2, {3}]],
      Joined→ False, PlotStyle→ Red]
  }, PlotRange→ All, ImageSize →500]
```



Nondipole contributions in a crossed-beam setup

This section explores the harmonic emission by a crossed-beam setup, with nondipole contributions, as a benchmarking step for the latter. The crossed-beam setup was proposed by X.-M. Tong and S.-I. Chu in *Phys. Rev. A* **58** no .4, R2656 (1998), and it was explored in a nondipole setting by V. Averbukh et al. in *Phys Rev. A* **65,** 063402 (2002). The results below reproduce those of Averbukh et al.

In short, we consider the harmonic emission by a circularly polarized pulse propagating along the *z* direction, at frequency ω, and a linearly polarized pulse of frequency $r\,\omega$ propagating along the *x* direction and polarized along the *z* direction.

```
(crossedBeamsA [x_, z_] = Function[t,
          {F1/ω Cos[k z - ω t], F1/ω Sin[k z - ω t], F2/(r ω) Sin[r k x - r ω t + θ0]}
        ])[t] // MatrixForm
(crossedBeamsGA [x_] = Function[t, Evaluate[{
              D[crossedBeamsA [x, z][t], x] /. {z → 0},
              D[crossedBeamsA [x, z][t], y] /. {z → 0},
              D[crossedBeamsA [x, z][t], z] /. {z → 0}
            }]]
    )[t] // MatrixForm
```

$$\begin{pmatrix} \frac{F1\,\mathrm{Cos}[k z - t\omega]}{\omega} \\ \frac{F1\,\mathrm{Sin}[k z - t\omega]}{\omega} \\ \frac{F2\,\mathrm{Sin}[k r x + \theta 0 - r t\omega]}{r\omega} \end{pmatrix}$$

$$\begin{pmatrix} 0 & 0 & \frac{F2\,k\,\mathrm{Cos}[k r x + \theta 0 - r t\omega]}{\omega} \\ 0 & 0 & 0 \\ \frac{F1\,k\,\mathrm{Sin}[t\omega]}{\omega} & \frac{F1\,k\,\mathrm{Cos}[t\omega]}{\omega} & 0 \end{pmatrix}$$

The dipole selection rules allow harmonic orders of the form $2r/ \pm 1$, with $/ = 0, 1, 2, 3, \ldots$, with polarization in the x, y plane, and harmonics of order $r(2/ + 1)$, with $/ = 0, 1, 2, 3, \ldots$, polarized along the z direction.

```
allowedHarmonics [r_, {1, 2}] := Select[Union[2 r Range[0, 100] + 1, 2 r Range[0, 100] - 1], # > 0 &]
allowedHarmonics [r_, {3}] := r (2 Range[0, 100] + 1)
```

For the calculation, then, some preliminaries,

```
αRange = {0, 1 / 137};
nppcb = 240;
crossedBeamsParameters  [rr_] := {F1 → 0.1, F2 → 0.2, ω → 0.057, θ0 → 0, r → rr, k → α ω};
DistributeDefinitions["RBSFA`"];
SetSharedFunction[crossedBeamsResults ];
```

and the calculation itself for $r = 2$ and $r = 5$.

```
Print[DateString[]]
ParallelTable[AbsoluteTiming [
    crossedBeamsResults [r, α] = makeDipoleList [
          VectorPotential→ crossedBeamsA [0, 0], VectorPotentialGradient→ crossedBeamsGA [0],
          FieldParameters → crossedBeamsParameters  [r]
          , DipoleTransitionMatrixElement→ Function[{p, κ}, gaussianDTME[p, 1 / 1.3]]
          , CarrierFrequency→ 0.057, PointsPerCycle→ nppcb
        ];
  ], {r, {2, 5}}, {α, αRange}]
Print[DateString[]]
```

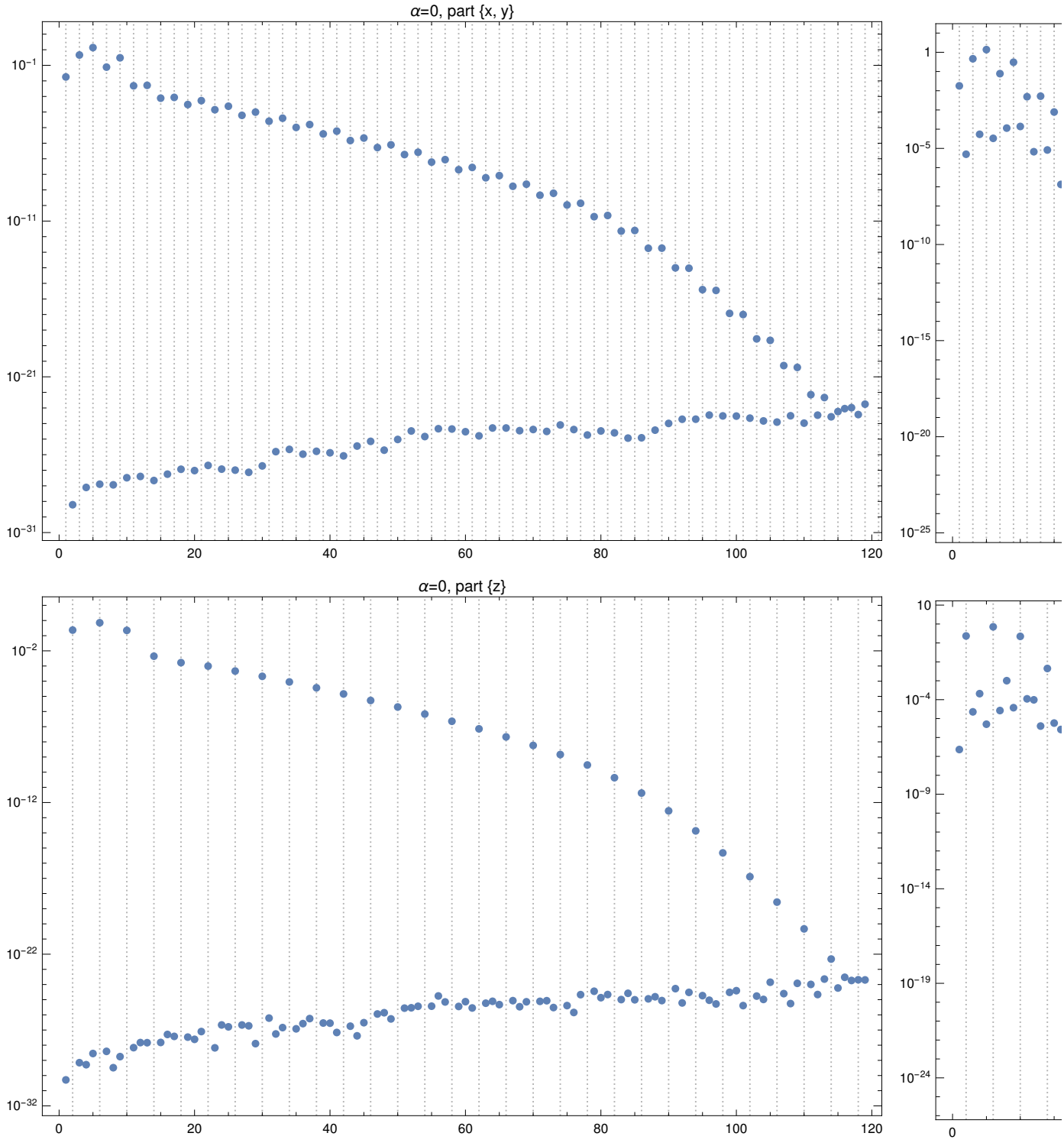
```
Thu 1 Oct 2015 15:42:15
```

```
{{{29.6518, Null}, {34.7323, Null}}, {{28.9947, Null}, {34.9678, Null}}}
```

```
Thu 1 Oct 2015 15:43:20
```

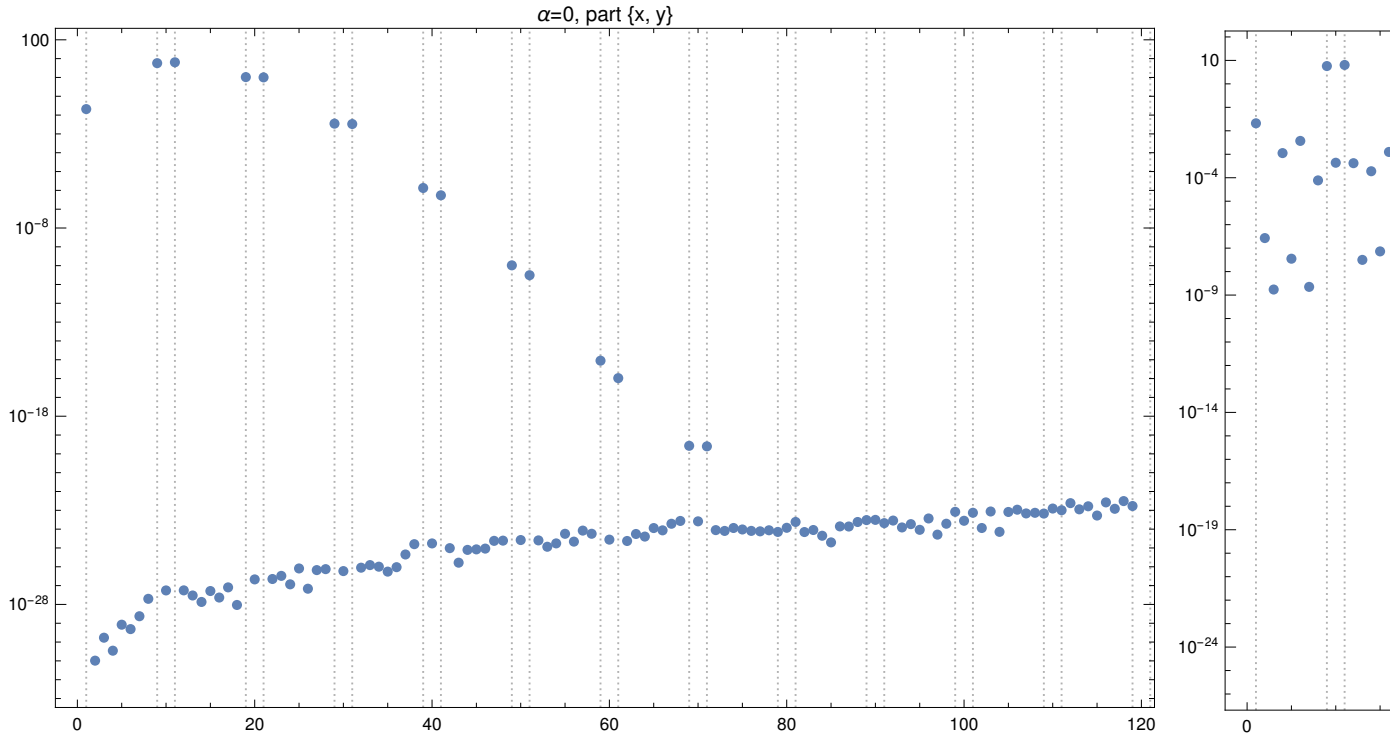
Results for $r = 2$, comparable to Fig. 1 in Averbukh et al. Dashed lines mark the dipole-allowed harmonics. The left-hand column has nondipole contributions turned off ($\alpha = 0$), and the right-hand column includes the nondipole contributions and observes a massive increase in the amplitude of the dipole-forbidden harmonics.

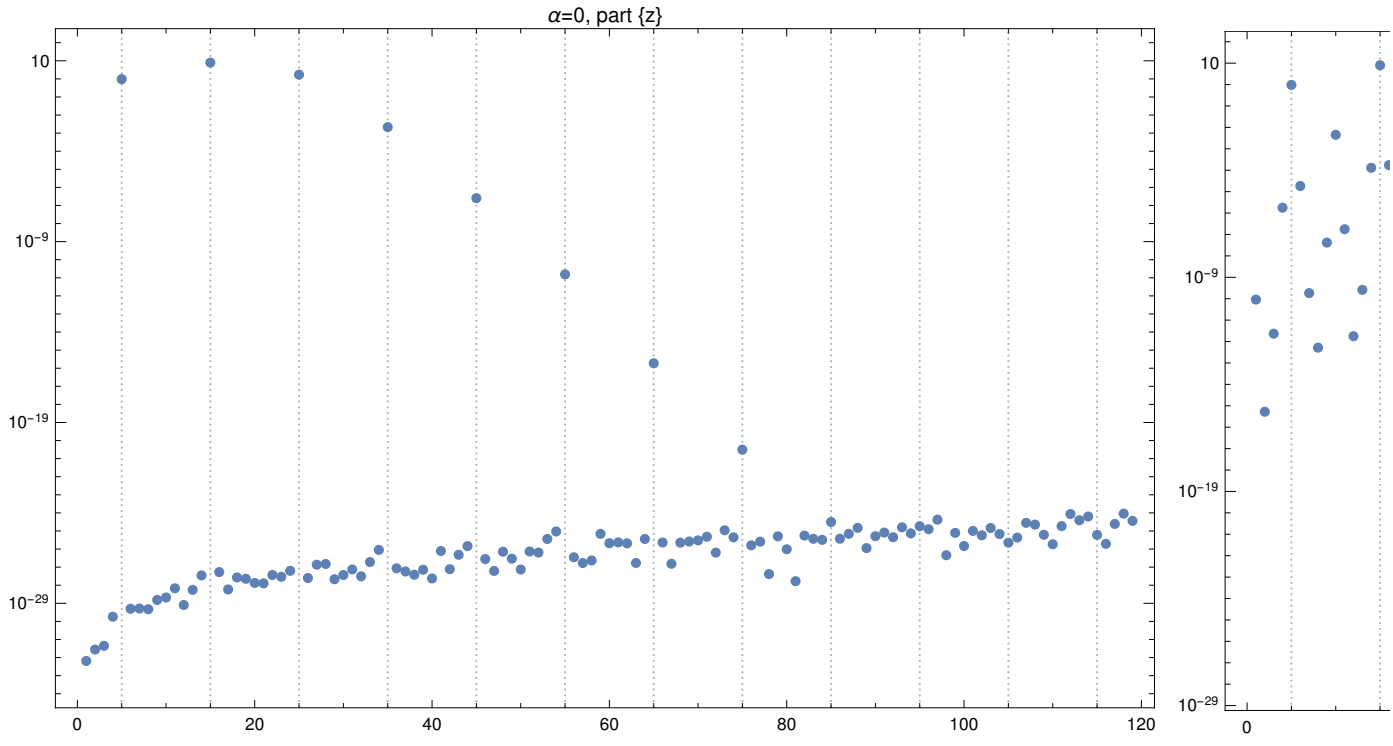
```
Grid[Table[
    ListLogPlot[
      getSpectrum [crossedBeamsResults [2, α][[1 ;; -2, part]], ωPower → 2][[2 ;;]]
      , Joined→ False, ImageSize → 600, PlotTheme → "Detailed", PlotRange→ Full
      , GridLines→ {allowedHarmonics [2, part], None}
      , PlotLabel → Row[{"α=", α, ", part ", {"x", "y", "z"}[[part]]}]
    ], {part, {{1, 2}, {3}}}, {α, αRange}]]
```



Results for $r = 5$, comparable to Fig. 2 in Averbukh et al.

```
Grid[Table[
    ListLogPlot[
      getSpectrum [crossedBeamsResults [5, α][[1 ;; -2, part]], ωPower → 2][[2 ;;]]
      , Joined→ False, ImageSize → 600, PlotTheme → "Detailed", PlotRange→ Full
      , GridLines→ {allowedHarmonics [5, part], None}
      , PlotLabel → Row[{"α=", α, ", part ", {"x", "y", "z"}[[part]]}]
    ], {part, {{1, 2}, {3}}}, {α, αRange}]]
```





Debugging and benchmarking tools

If something goes funny with your calls, then before you start taking makeDipoleList apart you can try using its Verbose option to diagnose the internal functions it is using. In particular:

- Setting Verbose→1 makes makeDipoleList print the Information of the key internal functions it is using, before it goes on to the integration loop.

```
makeDipoleList [VectorPotential→ Function[t, {F/ω Sin[ωt], 0, 0}],
    FieldParameters →{F→0.05, ω→0.057}, Verbose→1][[1 ;; 10]]
```

```
RBSFA`Private`ps
```

```
RBSFA`Private`ps[RBSFA`Private`t_?RBSFA`Private`gridPointQ
    RBSFA`Private`tt_?RBSFA`Private`gridPointQ] :=
  RBSFA`Private`ps[RBSFA`Private`t, RBSFA`Private`tt] = -1/(RBSFA`Private`t-RBSFA`Private`tt-i RBSFA`Private`ϵ)
      Inverse[IdentityMatrix[Length[RBSFA`Private`A[RBSFA`Private`tInit]]] -
        (RBSFA`Private`GAIntInt[RBSFA`Private`t, RBSFA`Private`tt] +
          Transpose[RBSFA`Private`GAIntInt[RBSFA`Private`t, RBSFA`Private`tt]]) /
        (RBSFA`Private`t-RBSFA`Private`tt-i RBSFA`Private`ϵ)].
      (RBSFA`Private`AInt[RBSFA`Private`t, RBSFA`Private`tt] -
        RBSFA`Private`bigPScorrectionInt[RBSFA`Private`t, RBSFA`Private`tt])
```

```
RBSFA`Private`pi
```

```
RBSFA`Private`pi[RBSFA`Private`p_, RBSFA`Private`t_] :=
  RBSFA`Private`p+RBSFA`Private`A[RBSFA`Private`t] -
    RBSFA`Private`GAInt[RBSFA`Private`t].RBSFA`Private`p-RBSFA`Private`GAdotAInt[RBSFA`Private`t]
```

```
RBSFA`Private`S
```

```
RBSFA`Private`S[RBSFA`Private`t_?RBSFA`Private`gridPointQ
    RBSFA`Private`tt_?RBSFA`Private`gridPointQ] :=
  1/2 (Norm [RBSFA`Private`ps[RBSFA`Private`t, RBSFA`Private`tt]]^2+RBSFA`Private`κ^2)
      (RBSFA`Private`t-RBSFA`Private`tt)+RBSFA`Private`ps[RBSFA`Private`t, RBSFA`Private`tt].
      RBSFA`Private`AInt[RBSFA`Private`t, RBSFA`Private`tt] +
    1/2 RBSFA`Private`A2Int[RBSFA`Private`t, RBSFA`Private`tt] -
    (RBSFA`Private`ps[RBSFA`Private`t, RBSFA`Private`tt].RBSFA`Private`GAIntInt[RBSFA`Private`t,
        RBSFA`Private`tt].RBSFA`Private`ps[RBSFA`Private`t, RBSFA`Private`tt] +
      RBSFA`Private`ps[RBSFA`Private`t, RBSFA`Private`tt].
        RBSFA`Private`bigPScorrectionInt[RBSFA`Private`t, RBSFA`Private`tt] +
      RBSFA`Private`AdotGAdotAInt[RBSFA`Private`t, RBSFA`Private`tt])
```

```
RBSFA`Private`AInt
```

```
RBSFA`Private`AInt[RBSFA`Private`t$_] = {-15.3894 Cos[0.057 RBSFA`Private`t$], 0, 0}

RBSFA`Private`AInt[RBSFA`Private`t$_, RBSFA`Private`tt$_] =
  {15.3894 Cos[0.057 RBSFA`Private`tt$] - 15.3894 Cos[0.057 RBSFA`Private`t$], 0, 0}
```

(abridged.)

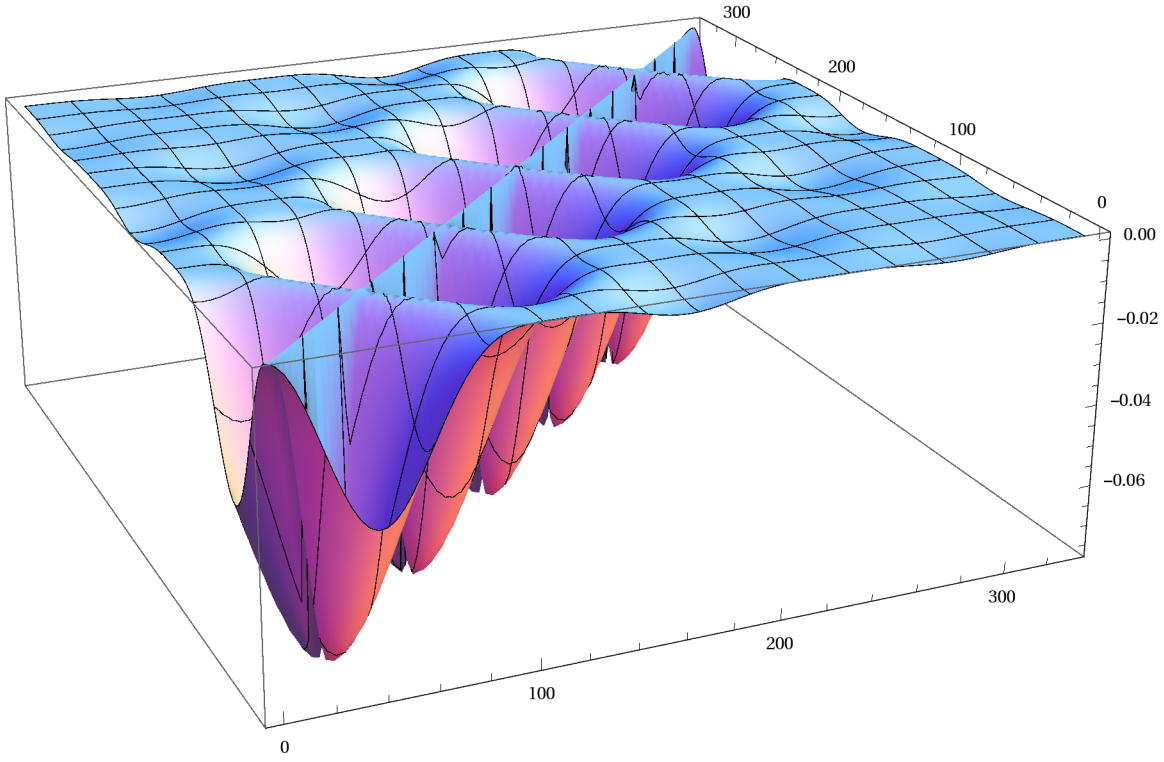
```
{{-0.0198736+0.00113629 i, 0.+0. i, 0.+0. i}, {-0.0166186-1.44349 i, 0.+0. i, 0.+0. i},
  {-0.0132498-5.34015 i, 0.+0. i, 0.+0. i}, {-0.00957477-10.5721 i, 0.+0. i, 0.+0. i},
  {-0.00563402-15.8027 i, 0.+0. i, 0.+0. i}, {-0.00185819-19.9418 i, 0.+0. i, 0.+0. i},
  {0.00123386-22.4066 i, 0.+0. i, 0.+0. i}, {0.00353492-23.1295 i, 0.+0. i, 0.+0. i},
  {0.0053967-22.4 i, 0.+0. i, 0.+0. i}, {0.00707192-20.6646 i, 0.+0. i, 0.+0. i}}
```

- Setting Verbose→2 makes makeDipoleList output its key internal functions and shut down before the integration takes place. Its results can be caught as follows:

```
{ps[t_, tt_], pi[p_, t_], S[t_, tt_], AInt[t_], AInt[t_, tt_], A2Int[t_], A2Int[t_, tt_],
    GAInt[t_], GAInt[t_, tt_], GAdotAInt[t_], GAdotAInt[t_, tt_], AdotGAInt[t_],
    AdotGAInt[t_, tt_], GAIntInt[t_], GAIntInt[t_, tt_], bigPScorrectionInt[t_],
    bigPScorrectionInt[t_, tt_], AdotGAdotAInt[t_], AdotGAdotAInt[t_, tt_], integrand[t_, τ_]
  } = makeDipoleList [VectorPotential→ Function[t, {F/ω Sin[ω t], 0, 0}],
    FieldParameters →{F→0.05, ω→0.057}, Verbose→2];
```

This then enables examination of e.g. the action:

```
Block[{ω = 0.057},
  Plot3D[
    Im [S[t, tt]]
    , {t, 0, 3 2π/ω}, {tt, 0, 3 2π/ω}
    , PlotRange→Full, ImageSize →600, PlotTheme →"Classic", PlotPoints→100
  ]
]
```



See the implementation notes in the code for makeDipoleList for further definitions of what each term entails.

Bicircular fields

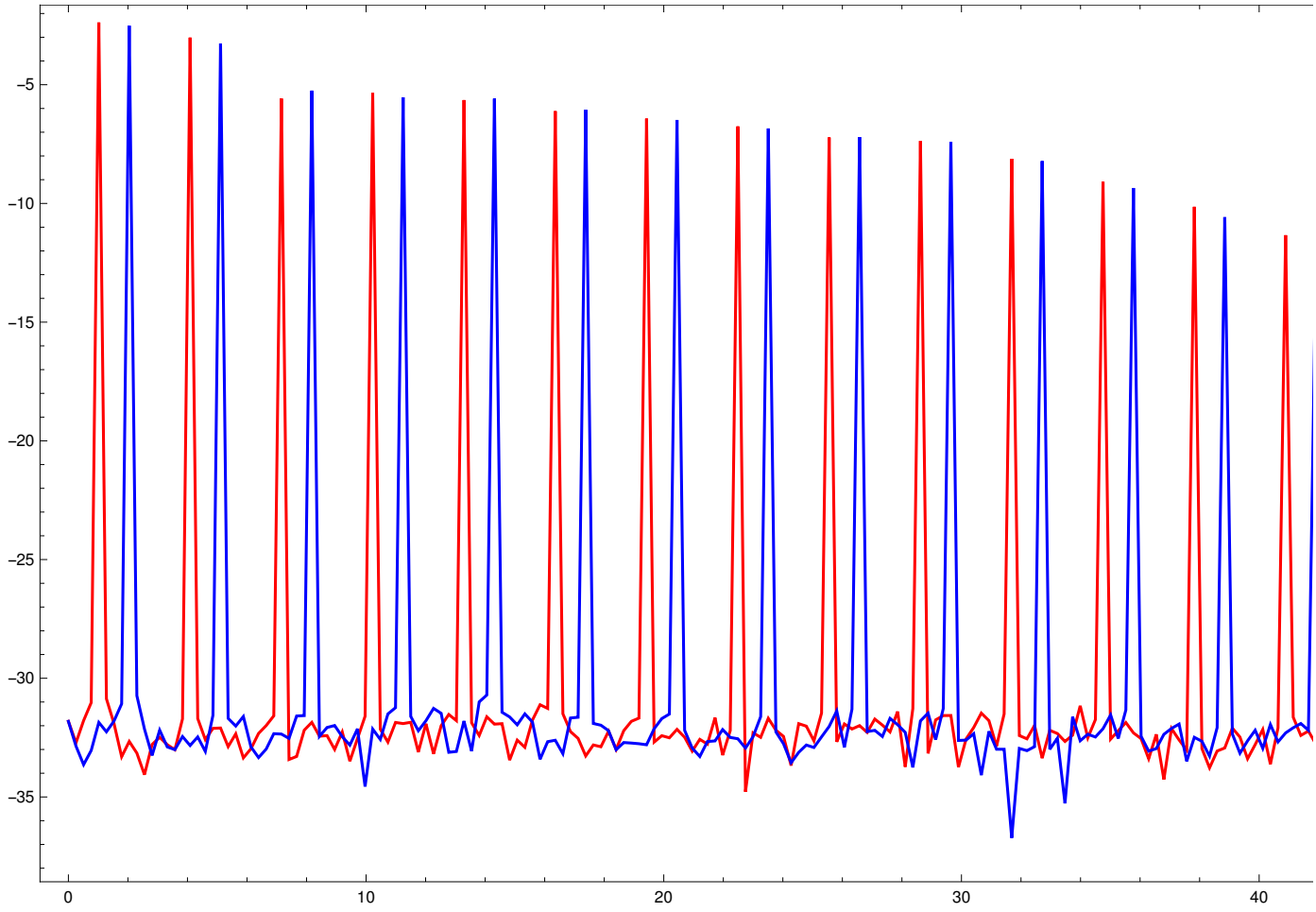
As a slightly less trivial example, consider a bicircular field: two counter-rotating, circularly polarized fields of different frequencies. The 'standard' case - as first demonstrated experimentally - has one field as the second harmonic of the fundamental, with both at equal intensities. The resultant harmonics appear at all integer orders except those divisible by three, with the $3n+1$ harmonics polarized as the fundamental, and the $3n-1$ harmonics polarized as the second-harmonic driver.

```
bicircularA[t_] := (F1/ω1 {Cos[t ω1] Sin[α], -Cos[α] Sin[t ω1]} + F2/ω2 {Cos[β] Cos[ω2 t], Sin[β] Sin[ω2 t]})
bicircularParameters = {F1 → 0.075, F2 → 0.075, α → 45°, β → 45°, ω1 → 45.6/800, ω2 → 45.6/400};
AbsoluteTiming [bicircularTest= makeDipoleList [VectorPotential→bicircularA,
        FieldParameters →bicircularParameters , TotalCycles→4];]
```

```
{8.45739, Null}
```

The function biColorSpectrum takes the spectrum and plots it, separating the two circular polarizations into different colours.

```
biColorSpectrum [Most[bicircularTest]]
```



Bicircular fields with a sine-squared envelope

To benchmark the original calculations, we compared them with the output of full MCTDH calculations. Here we used a $\sin^2$ envelope as the TDSE numerics require a finite pulse; the calculations take correspondingly longer but

they are still very manageable (two/three minutes per calculation for a fifteen-cycle pulse, resolving up to ~70 harmonics). One distinctive feature is that the harmonics near the cutoff are broader, because less cycles contribute to those energies.

```
bicircularEnvelopeA[t_] := cosPowerFlatTop[ω1, TotalCycles, 2][t]
      (F1/ω1 {Cos[t ω1] Sin[α], -Cos[α] Sin[t ω1]} + F2/ω2 {Cos[β] Cos[ω2 t], Sin[β] Sin[ω2 t]});
bicircularParameters = {F1 → 0.075, F2 → 0.075, α → 45°, β → 45°, ω1 → 45.6/800, ω2 → 45.6/400};
```

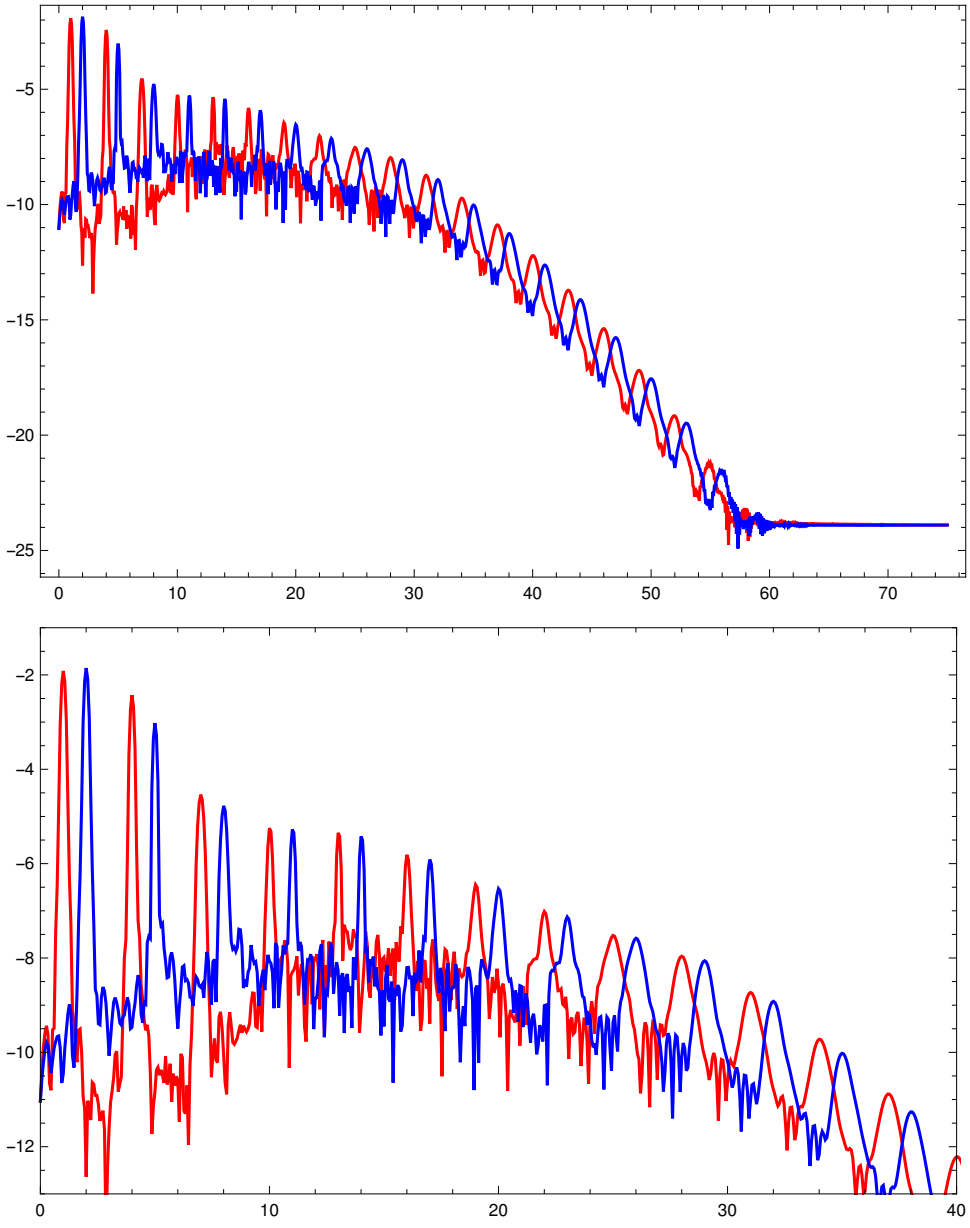
If (as in this case) the field depends on a number-of-cycles parameter, care must be taken that it matches the num option of the main call.

```
AbsoluteTiming [
  bicircularEnvelopeDipole = makeDipoleList [VectorPotential → bicircularEnvelopeA,
       FieldParameters → Join[bicircularParameters, {TotalCycles → 15}],
       PointsPerCycle → 150, TotalCycles → 15];
]
```

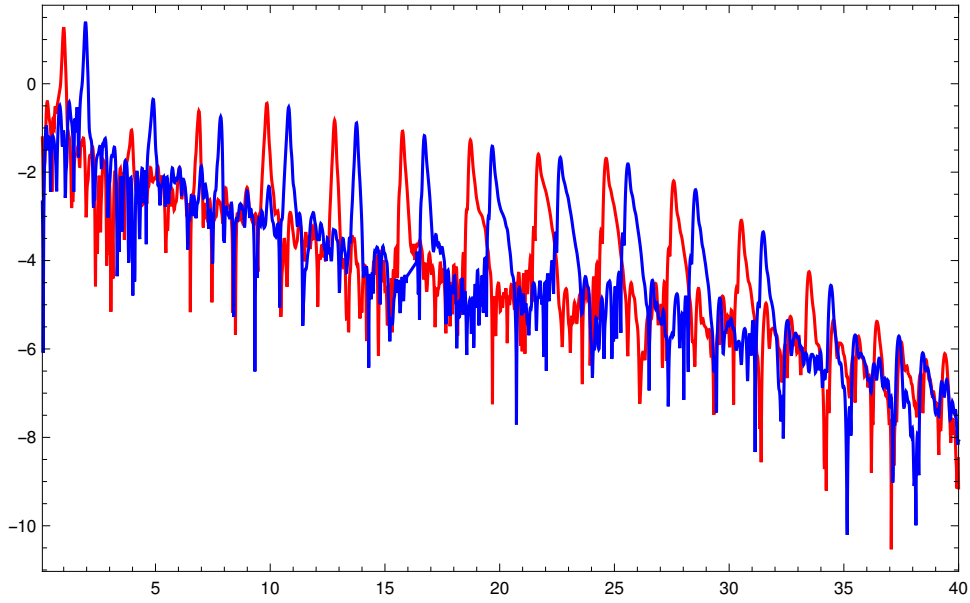
```
{141.302, Null}
```

Plotting the spectrum, and a zoom at the plateau:

```
biColorSpectrum [bicircularEnvelopeDipole,
  PointsPerCycle→150, TotalCycles→15, ImageSize →500]
biColorSpectrum [bicircularEnvelopeDipole, PointsPerCycle→150,
  TotalCycles→15, ImageSize →500, PlotRange→{{0, 40}, {-13, -1}}]
```



The comparable MCTDH spectrum, for identical conditions, looks like this:



Original RB-SFA: 'rotating' bicircular fields

Calculation

Here the fundamental laser driver has been set at an elliptical polarization (as in the original experiment, A. Fleischer et al., *Nature Photon.* **8**, 543 (2014)), which helps investigate the spin-angular-momentum conservation properties of HHG. In the model proposed in the original paper (*Phys. Rev. A* **90**, 043829 (2014)), the photon model is validated by splitting the elliptical field itself into two circular components, which can then be tuned independently:

$$\text{rotatingBicircularA}[t\_] := \text{envelope}[t]\left(\frac{F2}{\omega 2}\{\text{Cos}[\beta]\,\text{Cos}[\omega 2\,t-\phi 1],\ \text{Sin}[\beta]\,\text{Sin}[\omega 2\,t-\phi 1]\}+\frac{F1}{\sqrt{2}}\left(\frac{1}{\omega 1}\text{Cos}\left[\alpha-\frac{\pi}{4}\right]\{\text{Cos}[\omega 1\,t+\phi 1],\ -\text{Sin}[\omega 1\,t+\phi 1]\}+\frac{1}{(1+\delta)\,\omega 1}\text{Sin}\left[\alpha-\frac{\pi}{4}\right]\{\text{Cos}[(1+\delta)\,\omega 1\,t-\phi 1+\phi 2],\ +\text{Sin}[(1+\delta)\,\omega 1\,t-\phi 1+\phi 2]\}\right)\right);$$

```
DistributeDefinitions["RBSFA`"];
directory= FileNameJoin [{NotebookDirectory[], "Temp  Data"}];
filename [δ_] :=
    FileNameJoin [{directory, "data 25.09 detuning scan at δ=" <>ToString[δ] <>".txt"}];
Length[δRange = Range[0, 0.25, 0.001]]
```

```
251
```

To test the validity of the photon model, we ran a scan over the detuning δ, using the calculation below.

```
DateString[]
Print["Total = ", Length[δRange], " points at ~230s/point will be done at approximately  ",
  DateString[AbsoluteTime []+Length[δRange]*230./7], "."]
ParallelTable[
    Print[AbsoluteTiming [
        makeDipoleList [
            VectorPotential→rotatingBicircularA
            FieldParameters →{α→35°, β→45°, F1→0.075, F2→0.075, ω1→0.057,
                ω2→1.95×0.057, ϕ1→0, ϕ2→0, envelope→flatTopEnvelope[ω1, 26, 3]},
            CarrierFrequency→0.057, TotalCycles→26, PointsPerCycle→115,
            nGate→1.8, PointNumberCorrection →1, Preintegrals→"Numeric ",
            ReportingFunction→Function[Write[filename [δ], #]]
          ];]];
    Print[DateString[]];
    , {δ, δRange}];
DateString[]
NotebookSave[]
```

Total time 2h 32min. (Desktop machine with 8-thread, 4-core Intel i7-3770 CPU at 3.40GHz, 16GB RAB, running 7 *Mathematica* kernels in parallel.)
Expand this cell to see the calculation log.

The results can be pulled in from the files using this:

```
Do[detunedDipole[δ] = ReadList[filename [δ]], {δ, δRange}]
```

Or saved into a single location using this:

```
Save[FileNameJoin [{NotebookDirectory[], "Detuning scan collected data.txt"}], detunedDipole]
```

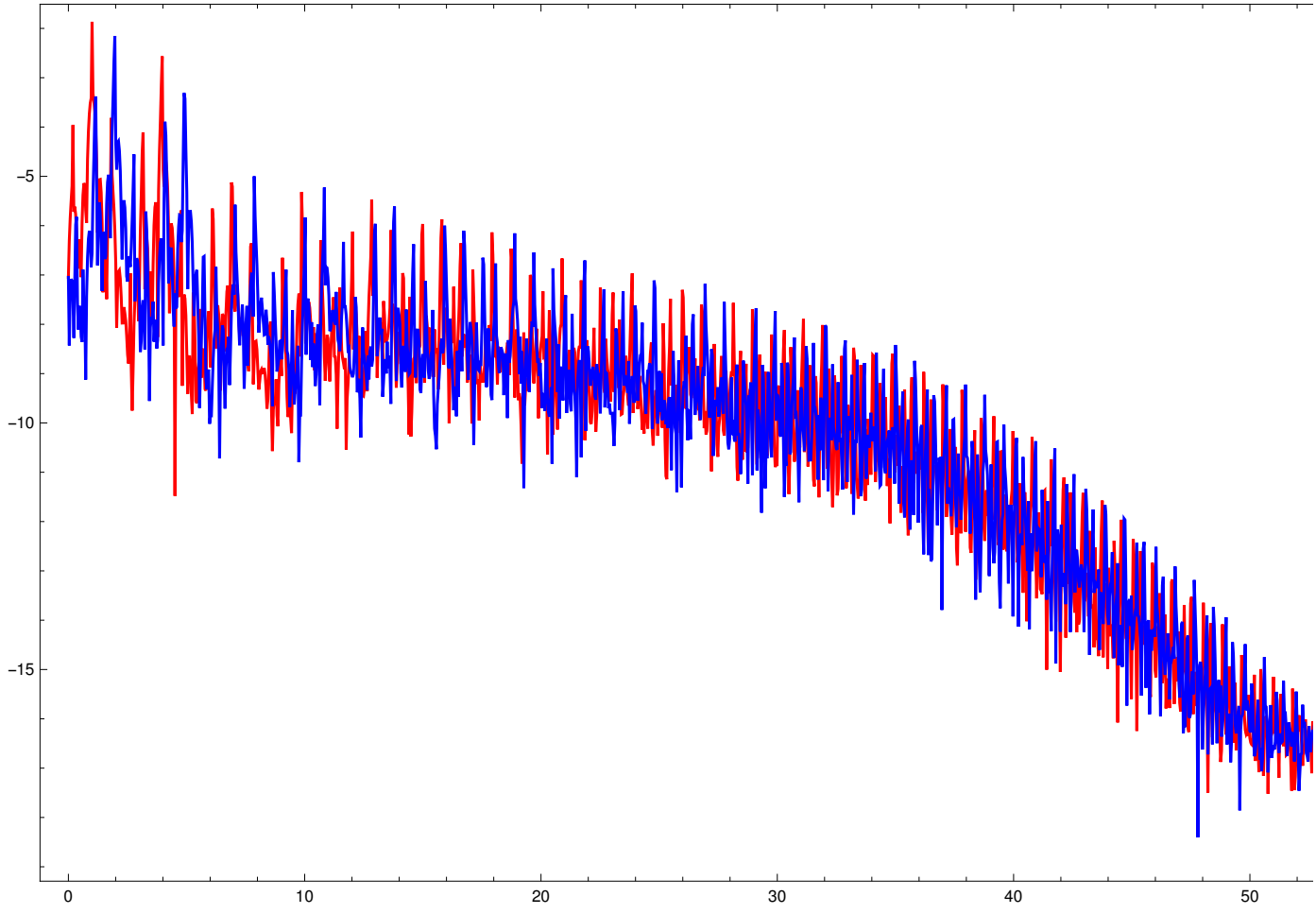
```
DumpSave [
    FileNameJoin [{NotebookDirectory[], "Detuning scan collected data.mx "}], detunedDipole];
```

and pulled in from the single location using this:

```
<< (FileNameJoin [{NotebookDirectory[], "Detuning scan collected data.txt"}]);
```

A sample spectrum looks like this:

```
biColorSpectrum [detunedDipole[0.001 RandomInteger [{0, 1000}]],
  CarrierFrequency→45.6/800, TotalCycles→3, PointsPerCycle→115]
```



Plots from the original paper

The plots from the original paper were produced using the code below. For simplicity we pre-define an interpolation function.

```
conditions:= Sequence[CarrierFrequency→45.6/800, TotalCycles→26, PointsPerCycle→115]
```

```
Remove [detuningInterpolation]
With[{length = Length[getSpectrum [detunedDipole[0.], Polarization→ {1, i}]]},
  AbsoluteTiming [
    Table[
        detuningInterpolation[ϵ] = Interpolation[
            Flatten[Table[
                {{
                        harmonicOrderAxis [TargetLength → length, conditions],
                        Table[δ, {length}]
                    }ᵀ,
                Log[10, getSpectrum [detunedDipole[δ], Polarization→ {1, ϵ i}]]
              }ᵀ
            , {δ, δRange}], 1]]
        , {ϵ, {1, -1}}];
  ]
]
{2.99829, Null}
```

Some plotting admin:

```
CMRmap = Function[x, Blend[{■, ■, ■, ■, ■, ■, ■, ■, □}, x]];
CMRwithMin[minIn_, minOut_: 1./9] :=
  Function[x, CMRmap [If[x < minIn, minOut/minIn x, minOut + (1 - minOut) (x - minIn)/(1 - minIn)]]]
min = 6. × 10^-9;
max = 5. × 10^-7;
colorfunction = CMRwithMin[min / max];
HOTicks[ϵ_] :=
  ({#, If[ϵ == 1, Style[#, Black], ""], {0.02, 0}, {Thickness[0.005], Gray}} & /@ Range[12, 18, 1]) ~
    Join~ ({#, "", {0.01, 0}, {Thickness[0.004], Gray}} & /@ Range[11 + 1/2, 18 + 1/4, 1/4])
downTicks = {{0, Style[0, Black], 0}, {0.25, Style[0.25, Black], 0}} ~Join~
      ({#, Style[#, Black], {0.015, 0}, {Thickness[0.005], Gray}} & /@ Range[0.05, 0.20, 0.05]) ~
      Join~ ({#, "", {0.01, 0}, {Thickness[0.004], Gray}} & /@ Range[0.01, 0.24, 0.01]);
upTicks = ({#, "", {0.015, 0}, {Thickness[0.005], Gray}} & /@ Range[0.05, 0.20, 0.05]) ~
      Join~ ({#, "", {0.01, 0}, {Thickness[0.004], Gray}} & /@ Range[0.01, 0.24, 0.01]);
```

The plot itself:

```
Row[Table[
    splittingsScan[ϵ] = RegionPlot[
        True
        , {δ, 0, 0.25}, {HO, 11.25, 18.5}
        , AspectRatio→ 1.2
        , PlotRangePadding→ None
        , ImagePadding →1 {{35+15 ϵ, 20}, {70, 6}}
        , ImageSize →{Automatic , 550}
        , PlotPoints→ 600
        , FrameStyle →Automatic
        , FrameLabel →
          {Style["ω'/ω-1", Black, 12], If[ϵ == 1, Style["Harmonic  Order", Black, 16], ""]}
        , ColorFunctionScaling→ False
        , FrameTicks →{{HOTicks[1], HOTicks[-1]}, {downTicks, upTicks}}
        , ColorFunction→ Function[{δ, HO}, colorfunction[10^detuningInterpolation[ϵ][HO, δ]/max]]
        , PlotLabel →
          Style[StringJoin[ϵ /. {1 → "Right", -1 → "Left"}, "-circular harmonics "], Black, 16]
      ]
    , {ϵ, {1, -1}}]
]
```

(Removed to keep file size low.)