

## Supplementary Information: Double nutation cross polarization between heteronuclear spins in solids

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### 1 Examples of DONUT waveforms

#### 1.1 Amplitude

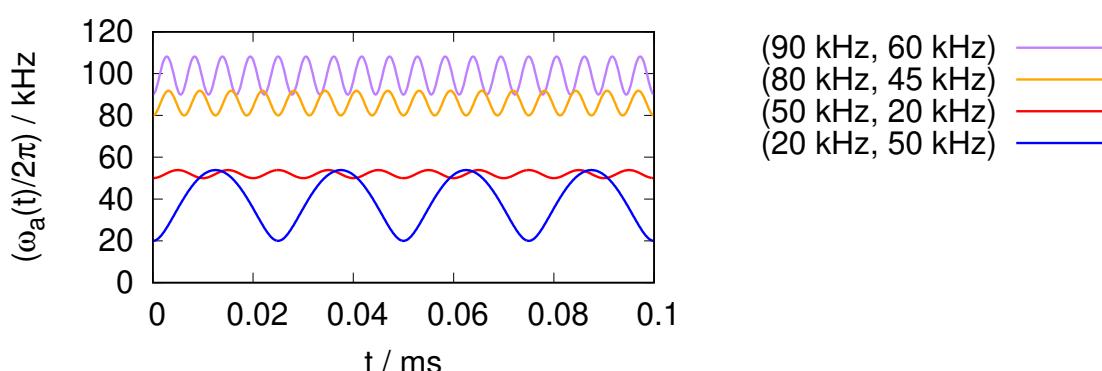


Figure S1: Profiles of the amplitude  $\omega_a(t)$  given in Eq. (32) for  $(\omega_{1I}/2\pi, \omega_{2I}/2\pi)$  of (20 kHz, 50 kHz), (50 kHz, 20 kHz), (80 kHz, 45 kHz), and (90 kHz, 60 kHz).

## 1.2 Phase

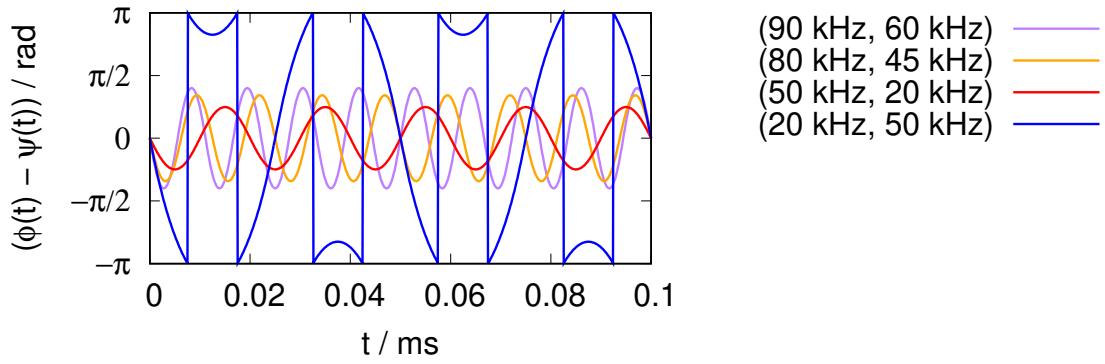


Figure S2: Profiles of the phase  $\phi(t) - \psi(t)$  given in Eqs. (33) and (42) for  $(\omega_{1I}/2\pi, \omega_{2I}/2\pi)$  of  $(20 \text{ kHz}, 50 \text{ kHz})$ ,  $(50 \text{ kHz}, 20 \text{ kHz})$ ,  $(80 \text{ kHz}, 45 \text{ kHz})$ , and  $(90 \text{ kHz}, 60 \text{ kHz})$ .

### 1.3 Gnuplot script for Fig. S1

```
set xrange [0:0.1]
set yrange [0:120]
set xl "t / ms"
set yl "(/Symbol w_a(t)/2/Symbol p) / kHz"
set key outside
set samples 1000
set size ratio 0.5
set terminal aqua size 640,480
# You may want to change "aqua" to the terminal type you prefer
f(nu1,nu2,x)=sqrt(nu1**2+nu2**2*sin(2*pi*nu1*x)**2)
plot f(90.0,60.0,x) lw 3 lc "purple" title "(90 kHz, 60 kHz)"
replot f(80.0,45.0,x) lw 3 lc "orange" title "(80 kHz, 45 kHz)"
replot f(50.0,20.0,x) lw 3 lc "red" title "(50 kHz, 20 kHz)"
replot f(20.0,50.0,x) lw 3 lc "blue" title "(20 kHz, 50 kHz)"
set terminal postscript eps color "Helvetica" 20
set output "amp.eps"
replot
```

### 1.4 Gnuplot script for Fig. S2

```
set xrange [0:0.1]
set yrange [-pi:pi]
set ytics ("‐p" ‐pi, "‐p/2" ‐pi/2, "0" 0.0, "p/2" pi/2, "p" pi) font "Symbol"
set xl "t / ms"
set yl "(/Symbol f(t) - /Symbol y(t)) / rad"
set key outside
set samples 1000
set size ratio 0.5
set terminal aqua size 640,480
# You may want to change "aqua" to the terminal type you prefer
f(nu1,nu2,x)=atan2(‐nu2*sin(2*pi*nu1*x),nu1)‐(nu2/nu1)*sin(2*pi*nu1*x)
plot atan2(sin(f(90.0,60.0,x)),cos(f(90.0,60.0,x))) lw 3 lc "purple" title "(90 kHz, 60 kHz)"
replot atan2(sin(f(80.0,45.0,x)),cos(f(80.0,45.0,x))) lw 3 lc "orange" title "(80 kHz, 45 kHz)"
replot atan2(sin(f(50.0,20.0,x)),cos(f(50.0,20.0,x))) lw 3 lc "red" title "(50 kHz, 20 kHz)"
replot atan2(sin(f(20.0,50.0,x)),cos(f(20.0,50.0,x))) lw 3 lc "blue" title "(20 kHz, 50 kHz)"
# IMPORTANT: ".0" is required to tell gnuplot that the values are float.
# 20/50 → 0!, while 20.0/50.0 → 0.4
set terminal postscript eps color "Helvetica" 20
set output "phase.eps"
replot
```