Supplemental material to the article "Second Optical Harmonics Generation Induced by Picosecond Terahertz Pulses in a Centrosymmetric Antiferromagnet NiO"

1. Visualization of the antiferromagnetic domains in NiO. In order to visualize the domain pattern in the studied sample we employed the optical polarimetry in the transmission geometry. In Fig. S1 c, d the images of the sample are shown as obtained in the polarization microscope. Given the polarization plane of the incoming light makes an angle 45° with the projection of **L** on the (001) plane, than the 90° antiferromagnetic domains can be visualized via magnetic linear birefringence [1]. Indeed, in Fig. S1 d, where polarizer transmission axis (P) was making an angle of 45° with the crystallographic direction [110], one can clearly see light and dark areas, corresponding to different T-domains. If the polarizer transmission axis is along the [100] crystallographic axis, then the contrast between these domains vanishes (Fig. S1 c). In this configuration clear contrast between the T-domains and domain walls between them is seen (thick red lines in Fig. 1S c, d). Furthermore, along with the 90° domain walls the 180° domain walls are visualized (thin red lines in Fig. S1 c). Naturally, such pairs of domains do not give any contrast in Fig. S1 d.

Larger part of the sample was in the multidomain state analogous to the one, shown in Fig. S1 c, d. Antiferromagnetic domain size was of order of 10 μm and was varying across the sample, in agreement with the previous reports on the domain patters in single crystalline unannealed NiO [2]. Furthermore, there was an area of 200 μ size, which was in a single domain state.

2. Components of the nonlinear susceptibility tensor describing spontaneous magneto-dipole SHG. For the magnetic point group $2/m$ the following components of the nonlinear susceptibility $\hat{\chi}^{eem}_{(S)}$, which is the 3$^{\text{rd}}$ rank axial tensor, are allowed by symmetry [2, 3]:

$$\begin{aligned}
\chi^{eem}_{zxx(S)} &= a; \quad \chi^{eem}_{xxz(S)} = \chi^{eem}_{xzx(S)} = b; \\
\chi^{eem}_{zyy(S)} &= c; \quad \chi^{eem}_{yyz(S)} = \chi^{eem}_{yzy(S)} = d; \\
\chi^{eem}_{zzz(S)} &= f; \quad \chi^{eem}_{zxy(S)} = \chi^{eem}_{zyx(S)} = g; \\
\chi^{eem}_{xyz(S)} &= \chi^{eem}_{xzy(S)} = h; \quad \chi^{eem}_{yxz(S)} = \chi^{eem}_{yzx(S)} = m.
\end{aligned} \tag{1}$$

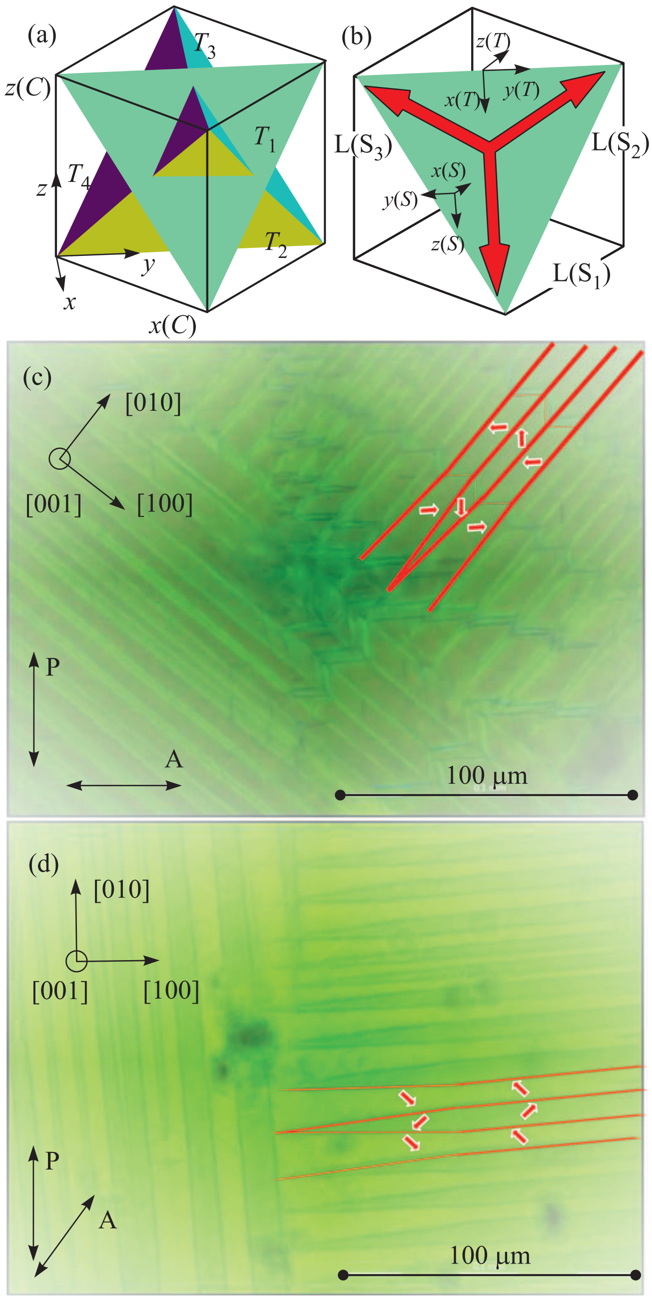

Fig. S1. (a) – Antiferromagnetic T- and (b) S-domains in a NiO crystal. (c), (d) – Images of the domain patter in the single crystalline 72 μm thick plate of NiO, as obtained in a polarization microscope for two orientations of the sample and analyzer. (c) – (100) axis makes an angle of 45° with the polarizer (P) axis, analyzer (A) and polarizer axes are crossed. (d) – (100) axis is parallel to the polarizer axis. Analyzer and polarizer axes make an angle of 45°. For the contrast enhancement the $\lambda/4$-plate was used in the geometry (d). Red arrows show projections of antiferromagnetic vector **L** from various T-domains on the (001) plane. Thick red lines mark 90°-walls between T-domains. Thin red lines mark 180°-walls between T-domains of the same type

Here the coordinate frame is associated with the S-domain ($\mathbf{z}(S)\|\mathbf{L}$) (Fig. S1 b). As was shown in a number of experiments on second harmonic generation (SHG) in magnetic crystals and summarized in [4], contributions to magnetic-dipole SHG due to tensor components of a type $\chi^{eem}_{xyz(S)}$ (g, h, and m in (1)) are negligible.

As discussed in details in [2], transition from the coordinate frame S associated with the S-domain to the laboratory coordinate frame L is realized via subsequent transformations

$$\begin{aligned} \chi^{eem}_{ijk} &= R(L \leftarrow S)_{ii'}R(L \leftarrow S)_{jj'}R(L \leftarrow S)_{kk'}\chi^{eem}_{ijk(S)}; \\ \hat{R}(L \leftarrow S) &= \hat{R}(L \leftarrow C)\hat{R}(C \leftarrow T)\hat{R}(T \leftarrow S), \end{aligned} \tag{2}$$

where $\hat{R}$ are the rotation matrices connecting laboratory coordinate frame, crystal axes (C), and coordinate frames associated with a T-domain (T; $\mathbf{z}(T)\|[111]$) and S-domain (see Fig. S1 a, b).

Applying there transformation one can find the components of the tensor $\hat{\chi}^{eem}$ in the laboratory coordinate frame, which describes the SHG process in the considered experimental geometry. In particular, for the S_1 domains belonging to various T-domains one finds:

$$\begin{aligned} \chi^{eem}_{xxy}(T_{1(3)}) &= \frac{2a}{3\sqrt{3}} + \frac{f}{3\sqrt{3}} - \frac{4b}{3\sqrt{3}}; \\ \chi^{eem}_{xyx}(T_{1(3)}) &= -\frac{2\sqrt{2}m}{3}; \\ \chi^{eem}_{xxy}(T_{2(4)}) &= \frac{f}{3} + \frac{b}{3\sqrt{3}}; \\ \chi^{eem}_{xyx}(T_{2(4)}) &= -\frac{\sqrt{2}f}{3} + \frac{\sqrt{2}b}{3\sqrt{3}}. \end{aligned} \tag{3}$$

Since $m(= \chi^{eem}_{yzx(S)})$ in this expression is negligible, one can expect the spontaneous second harmonic (SH) signal generated in the $T_{1(3)}$-domain to be polarized along the x-axis of the laboratory frame. In contrast, the SH signal polarized along the y-axis occurs in the $T_{2(4)}$-domain if the relation $b/f \approx -\sqrt{3}$, $a/b \approx 2 \pm \sqrt{2}$ holds. In fact, this situation was observed in our experiment (see Fig. 2a in the main text).

3. Components of the nonlinear susceptibility tensor describing electric-dipole SHG induced by external electric field. For the crystallographic point group $m3m$ there is electric field induced SHG of the electric-dipolar origin, which is described by the polar 4th rank tensor $\hat{\chi}^{eeee}_{(c)}$ [3]:

$$
\begin{aligned}
\chi^{eeee}_{iiii(C)} &= A\,(i = x, y, z); \\
\chi^{eeee}_{iijj(C)} &= B\,(i = x, y, z; j = x, y, z); \\
\chi^{eeee}_{ijij(C)} &= C\,(i = x, y, z; j = x, y, z); \\
\chi^{eeee}_{ijji(C)} &= D\,(i = x, y, z; j = x, y, z).
\end{aligned} \tag{4}
$$

Here the tensor components are written in the coordinate frame associated with the crystallographic axes. Using the transformation matrix $\hat{R}(L \leftarrow C)$ one can find the tensor components in the laboratory coordinate frame. For our experimental geometry only one component of the tensor $\hat{\chi}^{eeee}$ is relevant:

$$
\chi^{eeee}_{yxxy} = \frac{1}{2}(A - B). \tag{5}
$$

Thus, the THz-induced SHG should be polarized along the y-axis.

References

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