S1. Rate equation analysis for the NIR UC emission of Yb$^{3+}$/Tm$^{3+}$ codoped system

The mechanism of the NIR UC emission in Yb$^{3+}$/Tm$^{3+}$ codoped nanoparticles is depicted in Fig. S1. First, the Tm$^{3+}$ ion at state $^3H_6$ is excited to state $^3H_5$ through a phonon-assisted energy transfer from an excited Yb$^{3+}$ ion. Subsequently, the Tm$^{3+}$ ion relaxes non-radiatively to the lower state $^3F_4$ and is further excited to state $^3F_{2,3}$ through a second energy transfer process from excited Yb$^{3+}$ to the Tm$^{3+}$ ion. Finally, the Tm$^{3+}$ ion at state $^3F_{2,3}$ decays to state through an efficient non-radiative relaxation, and the NIR UC emission at 800 nm is generated by the transition of $^3H_4\rightarrow^3H_6$. 

S2. Rate equation analysis for the green UC emission of Yb$^{3+}$/Er$^{3+}$ codoped system

S3. Rate equation analysis for the green UC emission of Yb$^{3+}$/Ho$^{3+}$ codoped system

S4. The derivation of the analytical expression for the slope efficiency of the power-density dependence curve
Fig. S1  Schematic energy level diagrams of the Yb$^{3+}$ and Tm$^{3+}$ ions and the proposed UC mechanism following the excitation of 975-nm light.

The contribution of radiative rates to the depletion of states $^3H_5$ and $^3F_{2,3}$ are much less than their non-radiative decay rates, thus are omitted in the following discussion. Then the power-density dependent behavior of the NIR UC emission intensity under CW excitation can be described by the following steady rate equations:

\[
\frac{dN_{Yb1}}{dt} = \sigma \rho N_{Yb0} \frac{N_{Yb1}}{\tau_{Yb1}} = 0 ,
\]

\[
\frac{dN'_1}{dt} = C_0 N_0 N_{Yb1} - \beta'_1 N'_1 = 0 ,
\]

\[
\frac{dN_1}{dt} = \beta'_1 N'_1 - C_1 N_1 N_{Yb1} - \frac{N_1}{\tau_{1rad}} = 0 ,
\]

\[
\frac{dN'_2}{dt} = C_1 N_1 N_{Yb1} - \beta'_2 N'_2 = 0 ,
\]

\[
\frac{dN_2}{dt} = \beta'_2 N'_2 - \frac{N_2}{\tau_{2rad}} = 0 ,
\]

where $N_0$, $N_1$, $N'_1$, $N_2$ and $N'_2$ denote the population densities of the states $^3H_6$, $^3F_4$, $^3H_5$, $^3H_4$ and $^3F_{2,3}$ of Tm$^{3+}$ ions, respectively, while $N_{Yb0}$ and $N_{Yb1}$ are the population densities of the states $^2F_{7/2}$ and $^2F_{5/2}$ of Yb$^{3+}$ ions, respectively; $\sigma$ denotes the absorption cross-section of Yb$^{3+}$ ion; $\rho$ is the excitation photon flux, which is linearly related with power-density; $\tau_{1rad}$ and $\tau_{2rad}$ are the radiative lifetimes of Tm$^{3+}$ ions at states $^3F_4$ and $^3H_4$; $\tau_{Yb1}$ is the lifetime of Yb$^{3+}$ ions at $^2F_{5/2}$ state; $C_0$ and $C_1$ are ETU rates from excited Yb$^{3+}$ ions to the Tm$^{3+}$ ions at states $^3H_6$ and $^3F_4$, respectively; $\beta'_1$ and $\beta'_2$ represent the non-radiative decay rates for $^3H_5 \rightarrow ^3F_4$ and $^3F_{2,3} \rightarrow ^3H_4$, respectively. In this model, the depletion of the population density of the $^2F_{5/2}$ (Yb$^{3+}$) state due to ETU process is omitted, because the ETU rates at the $^2F_{5/2}$ (Yb$^{3+}$) state are much lower than its linear decay rate. For the same reason, the contribution to the depletion of state $^3H_4$ due to ETU to even higher states is not considered either. Under these assumptions, the population densities of different levels at steady state can be obtained from eqns (S1.1a) - (S1.1e)

\[
N_{Yb1} = \tau_{Yb1} \sigma N_{Yb0} \rho ,
\]

\[
N'_1 = \frac{C_0 N_0 N_{Yb1}}{\beta'_1} = \frac{C_0 N_0 N_{Yb1}}{\beta'_1} \sigma N_{Yb0} \rho ,
\]

\[
N_1 = \frac{C_1 N_1 N_{Yb1}}{\beta'_1} = \frac{C_1 N_1 N_{Yb1}}{\beta'_1} \sigma N_{Yb0} \rho .
\]
Thus, the power-density dependence of the UC steady-state emission from state $^3\text{H}_2$ has the form of

$$I = \frac{N_n^2}{\tau_2 \gamma} h \nu = \frac{C_0 C_1 N_0 (\sigma N_{\text{Yb}} \sigma N_{\text{Er}})^2}{\tau_1 \gamma + C_1 r_{\text{Yb}} \sigma N_{\text{Yb}} \rho}.$$  \hspace{1cm} (S1.3)

### S2. Rate equation analysis for the green UC emission of Yb$^{3+}$/Er$^{3+}$ codoped system

The mechanism of the NIR UC emission in Yb$^{3+}$/Er$^{3+}$ codoped nanoparticles is depicted in Fig. S2. As seen, the Er$^{3+}$ is promoted from the ground state $^4\text{I}_{15/2}$ to the $^4\text{F}_{7/2}$ state through two ETU processes from excited Yb$^{3+}$ ions. Then the Er$^{3+}$ ion relaxes non-radiatively to the lower states $^2\text{H}_{11/2}/^4\text{S}_{3/2}$, and generates green UC emissions through the transition of $^2\text{H}_{11/2}/^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$.

Fig. S2  Schematic energy level diagrams of the Yb$^{3+}$ and Er$^{3+}$ ions and the proposed UC mechanism following the excitation of 975-nm light.

The power-density dependent behavior of the green UC emission intensity under CW excitation can be described by the following rate equation model:

$$\frac{dN_{\text{Yb}}}{dt} = \sigma \rho N_{\text{Yb}0} - \frac{N_{\text{Yb}}}{\tau_{\text{Yb}}} = 0 \hspace{1cm} (S2.1a)$$

$$\frac{dN_1}{dt} = C_0 N_0 N_{\text{Yb}1} - C_1 N_1 N_{\text{Yb}1} - \beta_1 N_1 - \frac{N_1}{\tau_1} = 0 \hspace{1cm} (S2.1b)$$

$$\frac{dN_2}{dt} = C_1 N_1 N_{\text{Yb}1} - \beta_2 N_2 = 0 \hspace{1cm} (S2.1c)$$
where \( N_0, N_1, N_2 \) and \( N'_2 \) denote the population densities of the states \( ^4I_{15/2}, ^4I_{11/2}, ^2H_{11/2}^4S_{3/2} \) and \( ^2F_{7/2} \) of Er\(^{3+} \) ions, respectively, while \( N_{Yb0} \) and \( N_{Yb1} \) are the population densities of the states \( ^2F_{7/2} \) and \( ^2F_{5/2} \) of Yb\(^{3+} \) ions, respectively; \( \sigma \) denotes the absorption cross-section of Yb\(^{3+} \) ion; \( \rho \) is the excitation photon flux, which is linearly related with power-density; \( \tau_1^{\text{rad}} \) and \( \tau_2^{\text{rad}} \) are the radiative lifetimes of Er\(^{3+} \) ions at states \( ^4I_{11/2} \) and \( ^2H_{11/2}^4S_{3/2} \); \( \tau_{Yb1} \) is the lifetime of Yb\(^{3+} \) ions at \( ^2F_{7/2} \) state; \( C_0 \) and \( C_1 \) are ETU rates from excited Yb\(^{3+} \) ions to the Er\(^{3+} \) ions at states \( ^4I_{15/2} \) and \( ^4I_{11/2} \), respectively; \( \beta_1 \) and \( \beta_2 \) represent the non-radiative decay rate for \( ^4I_{11/2} \rightarrow ^4I_{13/2} \), \( ^4F_{7/2} \rightarrow ^2H_{11/2}^4S_{3/2} \), and \( ^2H_{11/2}^4S_{3/2} \rightarrow ^2F_{5/2} \), respectively.

In this model, the depletion of the population density of \( ^2F_{5/2} \) (Yb\(^{3+} \)) state due to ETU process is omitted, because the ETU rates at \( ^2F_{5/2} \) (Yb\(^{3+} \)) state are much lower than its linear decay rate. For the same reason, the contribution to the depletion of state \( ^2H_{11/2}^4S_{3/2} \) due to ETU to even higher states is not considered either. Under these assumptions, the population densities of different levels at steady state can be obtained from eqns (S2.1a) - (S2.1d)

\[
\frac{dN_2}{dt} = \beta_2'N'_2 - \beta_2N_2 - \frac{N_2}{\tau_2^{\text{rad}}} = 0, \quad (S1.1d)
\]

Let

\[
N_{Yb1} = \tau_{Yb1}\sigma N_{Yb0}\rho, \quad (S2.2a)
\]

\[
N_1 = \frac{C_0N_0N_{Yb1}}{\tau_1^{\text{rad}} + \beta_1 + C_1N_{Yb1}} = \frac{C_0N_0\tau_{Yb1}\sigma N_{Yb0}\rho}{\tau_1^{\text{rad}} + \beta_1 + C_1\tau_{Yb1}\sigma N_{Yb0}\rho}, \quad (S2.2b)
\]

\[
N'_2 = \frac{C_1N_1N_{Yb1}}{\beta_2} = \frac{C_0C_1N_0N_{Yb1}\sigma N_{Yb0}\rho}{\beta_2\left(\tau_1^{\text{rad}} + \frac{1}{\tau_1^{\text{rad}}} + \beta_1 + C_1\tau_{Yb1}\sigma N_{Yb0}\rho\right)}, \quad (S2.2c)
\]

\[
N_2 = \frac{\beta_2'N'_2}{\tau_2^{\text{rad}} + \beta_2} = \frac{C_0C_1N_0N_{Yb1}\sigma N_{Yb0}\rho}{\beta_2\left(\frac{1}{\tau_2^{\text{rad}}} + \frac{1}{\tau_1^{\text{rad}}} + \beta_1 + C_1\tau_{Yb1}\sigma N_{Yb0}\rho\right)}. \quad (S2.2d)
\]

Let

\[
\frac{1}{\tau_1^{\text{rad}}} + \beta_1 = \frac{1}{\tau_1}, \quad (S2.3)
\]

and

\[
\frac{1}{\tau_2^{\text{rad}}} + \beta_2 = \frac{1}{\tau_2}, \quad (S2.4)
\]

where \( \tau_1 \) and \( \tau_2 \) are the lifetimes of the state \( ^4I_{11/2} \) and \( ^2H_{11/2}^4S_{3/2} \) of Er\(^{3+} \), respectively, including the contribution of non-radiative decays, then we can obtain the power-density dependence of the green UC steady-state emission from the state \( ^2H_{11/2}^4S_{3/2} \)

\[
I = \frac{N_2}{\tau_2^{\text{rad}}} h\nu = \frac{C_0C_1\tau_{Yb1}(\tau_2^{\text{rad}})N_{Yb0}\rho\sigma^2 N_{Yb0}^2}{\tau_1 + C_1\tau_{Yb1}\sigma N_{Yb0}\rho}. \quad (S2.5)
\]

S3. Rate equation analysis for the green UC emission of Yb\(^{3+}/\)Ho\(^{3+} \) codoped system

The mechanism of the NIR UC emission in Yb\(^{3+}/\)Ho\(^{3+} \) codoped nanoparticles is depicted in Fig. S3. As seen, the Ho\(^{3+} \) at the ground state \( ^5I_8 \) is promoted to the \( ^3I_6 \) state through one energy transfer from excited Yb\(^{3+} \), and is further promoted to the \( ^5S_{2/2}^4F_{4} \) state through another energy transfer from the excited Yb\(^{3+} \) ions. The strong green UC emission can then be generated through the transition of \( ^5S_{2/2}^4F_{4} \rightarrow ^4I_6 \).
The power-density dependent behavior of the green UC emission intensity under CW excitation can be described by the following rate equation model:

\[
\frac{dN_{Yb1}}{dt} = \sigma \rho N_{Yb0} - \frac{N_{Yb1}}{\tau_{Yb1}} = 0, \quad (S3.1a)
\]

\[
\frac{dN_1}{dt} = C_0 N_0 N_{Yb1} - C_1 N_1 N_{Yb1} - \beta_1 N_1 - \frac{N_1}{\tau_1^{rad}} = 0, \quad (S3.1b)
\]

\[
\frac{dN_2}{dt} = C_1 N_1 N_{Yb1} - \beta_2 N_2 - \frac{N_2}{\tau_2^{rad}} = 0, \quad (S3.1c)
\]

where \(N_0, N_1, \) and \(N_2\) denote the population densities of the states \(^5I_8, ^5I_6, \) and \(^5S_{2/2}^F_4\) of Ho\(^{3+}\) ions, respectively, while \(N_{Yb0}\) and \(N_{Yb1}\) are the population densities of the states \(^2F_{7/2}\) and \(^2F_{5/2}\) of Yb\(^{3+}\) ions, respectively; \(\sigma\) denotes the absorption cross-section of Yb\(^{3+}\) ion; \(\rho\) is the excitation photon flux, which is linearly related with power-density; \(\tau_1^{rad}\) and \(\tau_2^{rad}\) are the radiative lifetimes of Ho\(^{3+}\) ions at states \(^5I_8\) and \(^5S_{2/2}^F_4\); \(\tau_{Yb1}\) is the lifetime of Yb\(^{3+}\) ions at \(^2F_{5/2}\) state; \(C_0\) and \(C_1\) are ETU rates from excited Yb\(^{3+}\) ions to the Ho\(^{3+}\) ions at states \(^5I_8\) and \(^5I_6\), respectively; \(\beta_1\) and \(\beta_2\) represents the non-radiative decay rate for \(^5I_8\rightarrow^5I_7\) and \(^5S_{2/2}^F_4\rightarrow^5F_5\), respectively. In this model, the depletion of the population density of \(^2F_{5/2}\) (Yb\(^{3+}\)) state due to ETU process is omitted, because the ETU rates at \(^2F_{5/2}\) (Yb\(^{3+}\)) state are much lower than its linear decay rate. For the same reason, the contribution to the depletion of state \(^5S_{2/2}^F_4\) due to ETU to even higher states is not considered either. Under these assumptions, the population densities of different levels at steady state can be obtained from eqns (S3.1a) - (S3.1c)

\[
N_{Yb1} = \tau_{Yb1} \sigma N_{Yb0} \rho, \quad (S3.2a)
\]

\[
N_1 = \frac{C_0 N_0 N_{Yb1}}{\frac{1}{\tau_1^{rad}} + \beta_1 + C_1 N_{Yb1}} = \frac{C_0 N_0 \tau_{Yb1} \sigma N_{Yb0} \rho}{\frac{1}{\tau_1^{rad}} + \beta_1 + C_1 \tau_{Yb1} \sigma N_{Yb0} \rho}, \quad (S3.2b)
\]

\[
N_2 = \frac{C_1 N_1 N_{Yb1}}{\frac{1}{\tau_2^{rad}} + \beta_2} = \frac{C_0 C_1 N_0 \tau_{Yb1} \sigma N_{Yb0} \rho}{\frac{1}{\tau_1^{rad}} + \beta_1 + C_1 \tau_{Yb1} \sigma N_{Yb0} \rho} + \beta_2, \quad (S3.2c)
\]
and

\[
\frac{1}{\tau_1^\text{rad}} + \beta_1 = \frac{1}{\tau_1}, \quad (S3.3)
\]

where \( \tau_1 \) and \( \tau_2 \) are the lifetimes of the state \(^5I_6\) and \(^5S_{2}/^5F_4\) of Ho\(^{3+}\), respectively, including the contribution of non-radiative decays, then we can obtain the power-density dependence of the green UC steady-state emission from the state \(^5S_{2}/^5F_4\)

\[
I = \frac{N_2}{\tau_2^\text{rad}} h\nu = \frac{C_0C_1\tau_{Yb1}^2(\tau_2/\tau_1^\text{rad})\eta_{0}h\nu\sigma^2N_{Yb0}\rho^2}{\tau_1 + C_1\tau_{Yb1}\sigma N_{Yb0}\rho}. \quad (S3.5)
\]

The NIR UC emission originating from Tm\(^{3+}\), the green UC emission from Er\(^{3+}\) and the green UC emission from Ho\(^{3+}\) have unified form of eqn (3). For the NIR UC emission of Tm\(^{3+}\),

\[
\tau_1 = \tau_1^\text{rad}. \quad (S3.6)
\]

and

\[
\tau_2 = \tau_2^\text{rad}. \quad (S3.7)
\]

**S4. The derivation of the analytical expression for the slope efficiency the power-density dependence curve**

The UC emission has a power-density dependence of

\[
I = \frac{a\rho^2}{b + c\rho}, \quad (S4.1)
\]

where

\[
a = C_0C_1\tau_{Yb1}^2(\tau_2/\tau_1^\text{rad})\eta_{0}h\nu\sigma^2N_{Yb0}^2, \quad (S4.2)
\]

\[
b = \frac{1}{\tau_1}, \quad (S4.3)
\]

and

\[
c = C_1\tau_{Yb1}\sigma N_{Yb0}. \quad (S4.4)
\]

Mathematically, the slope efficiency of the UC emission in a double-logarithmic scale is given by the derivative of \( \log I \) over \( \log \rho \), i.e.,

\[
k \equiv \frac{d\log I}{d\log \rho} = \frac{d\log I}{d\rho} \cdot \frac{d\rho}{d\log \rho} = \frac{d\log (\frac{a\rho^2}{b + c\rho})}{d\rho} \cdot \rho
\]
\[
\begin{align*}
\frac{a \rho^2}{b + c \rho} & \rightarrow \rho = \frac{2a \rho^2 (b + c \rho) - c a \rho^2}{(b + c \rho)^2}, \quad \rho \\
1 + \frac{1}{1 + \frac{c \rho}{b}} & = 1 + \frac{1}{1 + \Gamma_1^2 \Gamma Yb_1 \sigma N Yb_0 \rho} 
\end{align*}
\]