Effective lifetime of non-equilibrium carriers in semiconductors from non-adiabatic molecular dynamics simulations

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The lifetimes of non-equilibrium charge carriers in semiconductors calculated using non-adiabatic molecular dynamics often differ from experimental results by orders of magnitude. By revisiting the definition of carrier lifetime, we report a systematic procedure for calculating the effective carrier lifetime in semiconductor crystals under realistic conditions. The consideration of all recombination mechanisms and the use of appropriate carrier and defect densities are crucial to bridging the gap between modeling and measurements. Our calculated effective carrier lifetime of CH₃NH₃PbI₃ agrees with experiments, and is limited by band-to-band radiative recombination and Shockley–Read–Hall defect-assisted non-radiative recombination, whereas the band-to-band non-radiative recombination is found to be negligible. The procedure is further validated by application to the compound semiconductors CdTe and GaAs, and thus can be applied in carrier lifetime simulations in other material systems.

Excited-state carrier dynamics in semiconductors is fundamental to the development of optoelectronic, photovoltaic, photocatalytic and other functional devices working under light illumination. The lifetime of photo-excited non-equilibrium carriers (excess carriers generated by external excitation such as light illumination, which leads to deviation from thermodynamic equilibrium) is an important quantity in determining the performance of these devices. For example, it determines the diffusion length of minority carriers in solar cells. For decades, this important quantity has been obtained experimentally and other functional devices working under light illumination.

To determine the origin of the discrepancy we repeated the NAMD simulation. Our result agreed with theirs, so it appears that no technical error is causing the discrepancy. Kim and Walsh pointed out that non-adiabatic coupling of matrix elements between the valence and conduction bands may be significantly overestimated in NAMD simulations, resulting in exaggerated non-radiative recombination in pristine CH₃NH₃PbI₃ and thus leading to the short lifetime. This means that accuracy issues with current NAMD methods may be one possible origin of the discrepancy. On the other hand, as noted in refs. 7 and 8, most NAMD simulations use small supercells. The small supercell size constrains the simulation to have high values for the carrier density and defect density, which can differ from real conditions in experiments. These differences can also cause the observed discrepancy between computation and experiment. If the small-supercell-size effect is the origin, then two open questions need to be answered: (1) what is the physical meaning of the directly calculated carrier lifetime from NAMD simulations and (2) is it possible to use NAMD simulations to calculate a carrier lifetime that can be compared directly to the experimentally measured lifetime of real semiconductors working under light illumination?

In this Brief Communication we started from the fundamental definition of the lifetime of non-equilibrium carriers and developed a systematic procedure for calculating the effective carrier lifetime in real samples based on NAMD simulations, non-radiative...
multi-phonon theory and Fermi’s golden rule. Using this procedure, we clarified the origins of the discrepancy and determined the dominant recombination mechanisms in CH$_3$NH$_3$PbI$_3$ for computing an effective carrier lifetime consistent with experiments. The procedure was further validated on two compound semiconductors, GaAs and CdTe.

In the Methods we derive the procedure and formulae for calculating the effective lifetime $\tau$ of non-equilibrium carriers, including the influences of defect-assisted non-radiative Shockley–Read–Hall (SRH; $\tau_{SRH}$), band-to-band radiative ($\tau_{rad}$), band-to-band non-radiative ($\tau_{non-rad}^{band-band}$), and Auger ($\tau_{auger}$) recombinations. As shown in Fig. 1a, the SRH, band-to-band and Auger recombinations are one-carrier, two-carrier, and three-carrier processes, respectively, so their rates have linear, quadratic and cubic dependences on the density of non-equilibrium carriers $\Delta n(0)$ at the $t=0$ moment (when carrier generation stops), meaning that the dominant recombination mechanisms change as $\Delta n(0)$ changes.

Comparing the effective lifetime $\tau$ to the directly calculated lifetime from recent NAMD simulations$^{7,8}$, we find that recent studies considered only the influences of $\tau_{non-rad}^{band-band}$ and $\tau_{SRH}$. These simulations adopted supercells with several hundreds of atoms, and considered both defect-free supercells and those with defects. For defect-free supercells where no electronic states are found in the bandgap, band-to-band recombination is considered in the simulation, because the recombination occurs between electrons in the conduction band and holes (open orange circles) in the valence band; $e$ and $f$ represent the real semiconductor lattice space. The density of non-equilibrium carriers is only $10^{15}$ cm$^{-3}$ under sunlight illumination, as shown in $c$ and $e$, whereas the density imposed in NAMD simulation supercells can exceed $10^{20}$ cm$^{-3}$, as shown in $d$ and $f$, which can cause faster electron-hole recombination.

Fig. 1 | Calculating the carrier lifetime for three major recombination mechanisms. a, Schematic plot of first-order defect-assisted SRH, second-order band-to-band radiative or non-radiative, and third-order Auger recombination processes. $U$ represents the recombination rate. $A$, $B$ and $C$ are SRH, band-to-band and Auger recombination coefficients, respectively. $n_e$ and $p_h$ are the densities of electron and hole carriers under the equilibrium condition. $\Delta n$ is the density of non-equilibrium carriers. b, Systematic procedure for calculating the effective lifetime with the three major recombination mechanisms considered. $N_r$ is the density of the recombination-center defects and $E_r$ is the energy of the defect level. $n_e$ and $p_h$ are the densities of electron and hole carriers when the Fermi level is located at the defect level. $c_e$ and $c_p$ are the electron and hole capture coefficients of the defect level. $B_{rad}$ and $B_{non-rad}$ are the band-to-band radiative and non-radiative recombination coefficients. $\gamma_e$ and $\gamma_h$ are the e-e-h and h-h-e Auger recombination coefficients. $\tau_{SRH}$, $\tau_{band-band}$ and $\tau_{auger}$ are the lifetimes for SRH, band-to-band and Auger recombinations, respectively. $\tau_{eff}$ is the total effective carrier lifetime when the three recombination mechanisms are all considered. c-f, Comparison of band-to-band and SRH recombination processes in semiconductors with low and high densities of non-equilibrium carriers: $c$ and $d$ represent reciprocal band-structure space with electrons (shown by filled blue circles) in the conduction band and holes (open orange circles) in the valence band; $e$ and $f$ represent the real semiconductor lattice space. The density of non-equilibrium carriers is only $10^{15}$ cm$^{-3}$ under sunlight illumination, as shown in $c$ and $e$, whereas the density imposed in NAMD simulation supercells can exceed $10^{20}$ cm$^{-3}$, as shown in $d$ and $f$, which can cause faster electron-hole recombination.
recombination mechanisms (such as $\tau_{\text{rad-band-band}}$) are more important than $\tau_{\text{nond-rad-band}}$ and $\tau_{\text{SRH}}$ on $\Delta n(0)$ and $N_t$ (the density of recombination-center defects). In most NAMD simulations, the non-equilibrium electron and hole carriers are generated by exciting an electron from the VBM to the CBM. The supercell usually has several hundred atoms, so the density $\Delta n(0)$ of non-equilibrium carriers is as high as $10^{20}$ cm$^{-3}$ (Fig. 1d,f). For example, in a 192-atom CH$_3$NH$_3$PbI$_3$ supercell, the density of non-equilibrium carriers is $2.6 \times 10^{16}$ cm$^{-3}$. However, under sunlight illumination (Fig. 1c,e), the excited non-equilibrium carriers have a much lower density, $\sim 10^{15}$–$10^{17}$ cm$^{-3}$. The imposed $\Delta n(0)$ is therefore much higher than the density in real devices working under sunlight illumination, as compared in Fig. 1c–f. Similarly, the defect density $N_t$ is also seriously overestimated when calculating $\tau_{\text{SRH}}$.

Following equation (17) and Supplementary Section 1, $\tau_{\text{nond-rad-band}}$ is inversely proportional to $\Delta n(0)$ when $\Delta n(0)$ is high. Following equation (16) and Supplementary Section 2, $\tau_{\text{SRH}}$ is inversely proportional to $N_t$. Therefore, the exaggerated $\Delta n(0)$ and $N_t$ imposed in the several-hundred-atom supercells will cause the NAMD-simulated $\tau_{\text{nond-rad-band}}$ and $\tau_{\text{SRH}}$ to be seriously underestimated by orders of magnitude. The lifetime of $\sim 1.5$ ns estimated by NAMD simulations is much shorter than the measured lifetime in real samples with smaller values of $\Delta n(0)$ and $N_t$. In the Methods, we show the formula to convert the NAMD lifetime to the supercell-size-independent $\tau_{\text{nond-rad-band}}$ (band-to-band non-radiative recombination coefficient), $c_e$ and $c_h$ (electron and hole capture coefficients). These coefficients can be used to calculate the effective $\tau_{\text{nond-rad-band}}$ and $\tau_{\text{SRH}}$ in real semiconductors with realistic $\Delta n(0)$ and $N_t$, following the procedure in Fig. 1b. The calculations reveal that the effective $\tau_{\text{nond-rad-band}}$ and $\tau_{\text{SRH}}$ are indeed much longer and do not contradict the experimental lifetime.

Our analysis highlights two origins of the discrepancy between the NAMD-simulated and experimentally measured lifetimes: (1) imposition of high $\Delta n(0)$ and $N_t$ and (2) consideration of only $\tau_{\text{nond-rad-band}}$ and $\tau_{\text{SRH}}$ while neglecting $\tau_{\text{rad-band-band}}$ and $\tau_{\text{Auger}}$ and other recombination mechanisms. We now discuss the second point, that is, how the incomplete consideration of recombination mechanisms influences the total effective lifetime $\tau$ in CH$_3$NH$_3$PbI$_3$ under illumination.

Upon illumination, the non-equilibrium carrier density in CH$_3$NH$_3$PbI$_3$ increases and then approaches a steady state, as shown in Fig. 2a. The intensity of light illumination determines the carrier generation rate $G$. Under sunlight (one-sun) illumination, $G$ is $\sim 10^{14}$ cm$^{-3}$ s$^{-1}$. Under concentrated light, the light intensity becomes stronger, and thus $G$ increases. As $G$ increases, the steady-state carrier density can be increased (as shown in Fig. 2b), so the carrier lifetimes of different recombination mechanisms can be changed. Figure 2d–f shows the calculated $\tau_{\text{nond-rad-band}}$, $\tau_{\text{rad-band-band}}$, $\tau_{\text{SRH}}$ and $\tau_{\text{Auger}}$ as functions of $G$. The details of these calculations are provided in the Methods. $\tau_{\text{SRH}}$ is inversely proportional to the deep-level recombination-center defect density $N_t$. However, the defect density in real samples depends on the fabrication conditions and processes. To compare results for CH$_3$NH$_3$PbI$_3$ samples fabricated using different processes, Fig. 2d–f presents plots for three representative values of $N_t$: low at $10^{14}$ cm$^{-3}$, medium at $10^{15}$ cm$^{-3}$ and high at $10^{16}$ cm$^{-3}$.

Figure 2d shows the lifetime for a low $N_t$ of $10^{14}$ cm$^{-3}$. This low density is possible in CH$_3$NH$_3$PbI$_3$ because most of the deep-level defects were reported to have high formation energies and thus low densities under the equilibrium condition. The results showed that $\tau_{\text{SRH}}$ is always $\sim 0.1$ s, which is almost independent of $G$ because the influences of $\Delta n(0)$ on the SRH recombination coefficient $A$ are partially canceled in the numerator and denominator of equation (16). In contrast, $\tau_{\text{rad-band-band}}$ and $\tau_{\text{nond-rad-band}}$ are sensitive to $G$ and decrease quickly as the light intensity and $G$ increase, because $\tau_{\text{rad-band-band}}$ and $\tau_{\text{nond-rad-band}}$ are almost inversely proportional to $\Delta n(0)$ and thus also to $G$. Comparing $\tau_{\text{SRH}}$, $\tau_{\text{rad-band-band}}$, $\tau_{\text{nond-rad-band}}$ and $\tau_{\text{Auger}}$, band-to-band radiative recombination is the fastest recombination mechanism and determines the total effective lifetime $\tau$ when $G$ is lower than $10^{15}$ cm$^{-3}$ s$^{-1}$, and the Auger recombination dominates at very high $G$ (high carrier densities). In contrast, the SRH and band-to-band non-radiative mechanisms only cause slower recombination and thus just decrease $\tau$ slightly according to equations (7) and (20).

Upon sunlight illumination with $G$ of $10^{15}$ cm$^{-3}$ s$^{-1}$, the steady-state carrier density is $\sim 10^{16}$ cm$^{-3}$ when $N_t=10^{15}$ cm$^{-3}$ and thus the total effective lifetime $\tau \approx \tau_{\text{rad-band-band}} \approx 1 \mu$s. For $N_t$ in the range $10^{10}$–$10^{15}$ cm$^{-3}$, $\tau_{\text{SRH}}$ is always shorter than $\tau_{\text{SRH}}$ and the shortest among the lifetimes of all the recombination mechanisms under sunlight illumination. Therefore, $\tau_{\text{rad-band-band}}$ always determines the effective lifetime $\tau$ to be $\sim 1 \mu$s. The dominance of the band-to-band radiative recombination and the predicted long effective lifetime $\tau$ are consistent with the experimental finding of efficient photoluminescence and a measured long carrier lifetime of $\sim 1 \mu$s in CH$_3$NH$_3$PbI$_3$ single crystals with $N_t \approx 10^{15}$ cm$^{-3}$ (ref. 1). The consistency between the calculated and measured photoluminescence quantum yield (PLQY) is shown in Fig. 2c and discussed in Supplementary Section 3.

When $N_t$ increases, the dominant recombination mechanism changes. When $N_t$ increases to $10^{16}$ cm$^{-3}$, $\tau_{\text{SRH}}$ becomes only 10 ns (Fig. 2f). The SRH recombination dominates the total effective lifetime $\tau$ when the light intensity is low and $G$ is lower than $10^{14}$ cm$^{-3}$ s$^{-1}$, while the band-to-band radiative recombination dominates only in a small range of $G$ of $\sim 10^{14}$ cm$^{-3}$ s$^{-1}$. For even higher $G$, Auger recombination dominates. Under sunlight illumination with $G$ of $\sim 10^{16}$ cm$^{-3}$ s$^{-1}$, SRH recombination is dominant. CH$_3$NH$_3$PbI$_3$ polycrystals with a high $N_t$ of $10^{15}$–$10^{17}$ cm$^{-3}$ were reported to have carrier lifetimes in the range 0.01–1 $\mu$s (ref. 31), consistent with the calculated short $\tau_{\text{SRH}}$ for high $N_t$.

The above analysis confirms that the lifetime of non-equilibrium carriers in CH$_3$NH$_3$PbI$_3$ solar cells should be mainly determined by band-to-band radiative recombination and is usually very long. Only when thin films have a high density of recombination-center defects or dopants can SRH recombination appreciably decrease the lifetime. Across the whole range of $G$, $\tau_{\text{nond-rad-band}}$ is always longer than $\tau_{\text{rad-band-band}}$ by two orders of magnitude, so the influence of band-to-band non-radiative recombination on the effective lifetime $\tau$ is negligible in CH$_3$NH$_3$PbI$_3$, no matter whether it is under strong or weak light illumination. Therefore, changes in $\tau_{\text{nond-rad-band}}$ alone should not be over-interpreted, for example, 1.5 ns for the pristine CH$_3$NH$_3$PbI$_3$ compared to 4–11 ns for doped or defective crystals. The experimentally improved performance of CH$_3$NH$_3$PbI$_3$ solar cells after the alkaline-metal treatments should result from the substantially increased $\tau_{\text{SRH}}$ because the alkaline-metal dopants passivate the recombination-center level of the iodine interstitial and thus give a clean bandgap.

These results demonstrate that it is possible to calculate the carrier lifetime to agree with experimentally measured values, based on NAMD simulations. In Supplementary Section 4 and Extended Data Figs. 1 and 2, we further validate this on two compound semiconductors, CdTe and GaAs, with carrier lifetimes exceeding 1 $\mu$s for low defect densities. There are still limitations to the calculation procedure in Fig. 1b: (1) it considers only three major recombination mechanisms and does not consider the surface and interface recombination, which can be important and further shorten the effective carrier lifetime in polycrystalline and thin-film semiconductors.
(2) when there are several different types of recombination-center
defect in real samples, more than one SRH recombination pathway
exist and they should all be considered; (3) the approximations
to the exchange-correlation functional in the DFT and the many-
body effects can cause errors in the calculated band structure,
defect levels, electron–phonon coupling and thus the recombina-
tion coefficients, which can also limit the accuracy of the lifetime
calculation. These limitations should be noted when comparing
the calculated lifetime to the measured value of real samples.

Besides carrier recombination, other excited-state carrier-dynamics
processes are also widely simulated by NAMD- and TDDFT-related
methods, such as the laser-induced phase transition or melting,
radiolysis radiation damage, hot carrier cooling and photocurrent
generation in a strong external field. The timescales of these pro-
cesses are attracting great attention. Our finding of the factors influ-
encing the carrier recombination lifetime have implications for the
accurate calculations of these timescales. These processes may also
have several different mechanisms (or pathways), and their rates
may depend on the carrier density and thus on the supercell size
used in the simulation. Therefore, the consideration of all relevant

Fig. 2 | Calculated non-equilibrium carrier density, carrier lifetime and PLQY in CH$_3$NH$_3$PbI$_3$ under light illumination. a, Time evolution of the non-
equilibrium carrier density under sunlight illumination. b, Steady-state carrier density as a function of generation rate $G$ (light intensity). c, PLQY
dependence on steady-state carrier density. Experimental data are shown for comparison. d–f, Carrier lifetime as a function of $G$ for $N_t = 10^8$
(d), $10^{13}$ (e) and $10^{15}$ cm$^{-3}$ (f). The shaded areas show the ranges of $G$ with different dominant recombination mechanisms.
mechanisms and appropriate carrier densities also deserves attention in those studies.

**Methods**

*Definition of carrier lifetime and calculation procedure.* As introduced in semiconductor physics textbooks, the time derivative of the non-equilibrium carrier density $\Delta n(t)$ is equal to the difference between the generation rate $G$ and the recombination rate $U(t)$ of non-equilibrium carriers:

$$\frac{d\Delta n(t)}{dt} = G(t) - U(t).$$

When the generation stops, $G(t) = 0$, $\Delta n(t)$ will decay with the rate $-U(t)$. If the recombination rate $U(t)$ depends linearly on $\Delta n(t)$, that is

$$U(t) = \frac{\Delta n(t)}{\tau},$$

where $\tau$ is a constant,

$$\tau = \frac{\Delta n(t)}{U(t)},$$

then the decay of $\Delta n(t)$ will follow

$$\Delta n(t) = \Delta n(0) \exp\left(-\frac{U(t)}{\Delta n(t)}\right) = \Delta n(0) \exp\left(-\frac{t}{\tau}\right),$$

where $\Delta n(0)$ is the density of non-equilibrium carriers at the $t = 0$ moment when the generation stops. As we can see, in this case, the constant $\tau$ means the time when the density decays to $1/e$ of the original value $\Delta n(0)$ after the generation stops, so it is defined as the lifetime of non-equilibrium carriers. Then, equation (1) becomes

$$\frac{d\Delta n(t)}{dt} = -U(t) = -\frac{\Delta n(t)}{\tau}.$$ (5)

In standard NAMD studies, the lifetime $\tau$ is calculated by fitting the decay of the electron population on the excited-state level to the function $\Delta n(t) \exp(-t/\tau)$, which originates from the decay function in equation (4). According to equation (3), the lifetime $\tau$ is determined by the ratio between the density of non-equilibrium carriers $\Delta n(t)$ and the recombination rate $U(t)$. In semiconductors, as shown in Fig. 1a, there are many possible recombination mechanisms, for example, the defect-assisted non-radiative SRH recombination, the band-to-band recombination and the Auger recombination (for polycrystalline thin films, the surface and interface recombination may be important and should also be considered). Therefore, the recombination rate

$$U(t) = U_{\text{SRH}}(t) + U_{\text{band-band}}(t) + U_{\text{Auger}}(t)$$

is the sum of the rates of all the mechanisms. Correspondingly, the lifetime that is effective in real semiconductor samples should also be contributed by all the mechanisms:

$$\frac{1}{\tau} = \frac{1}{\tau_{\text{SRH}}} + \frac{1}{\tau_{\text{band-band}}} + \frac{1}{\tau_{\text{Auger}}},$$

where $\tau_{\text{SRH}}, \tau_{\text{band-band}}$ and $\tau_{\text{Auger}}$ are the lifetimes when only one of the three mechanisms is considered. With this definition, we can also write

$$\tau_{\text{SRH}} = \frac{\Delta n(t)}{U_{\text{SRH}}(t)},$$

$$\tau_{\text{band-band}} = \frac{\Delta n(t)}{U_{\text{band-band}}(t)},$$

$$\tau_{\text{Auger}} = \frac{\Delta n(t)}{U_{\text{Auger}}(t)}.$$ (10)

As shown by Grundmann, the three recombination rates depend not only on $\Delta n(t)$ (for photo-excited non-equilibrium carriers, the densities of non-equilibrium electrons and holes are equal, $\Delta n(t) = \Delta n(t)$), but also on the electron density $n_0$ and hole carrier density $p_0$ under the equilibrium state, as described by

$$U_{\text{SRH}}(t) = A \times \Delta n(t),$$

$$U_{\text{band-band}}(t) = B \times [n_0 + p_0 + \Delta n(t)] \times \Delta n(t),$$

$$U_{\text{Auger}}(t) = C \times \Delta n(t)^3,$$

in which, $A, B$ and $C$ are the SRH defect-assisted non-radiative, band-to-band and Auger recombination coefficients, respectively, $n_0$ and $p_0$ can be calculated as $n_0 = N_e \exp\left(\frac{E_{\text{CBM}} - E_{\text{C}}}{k_B T}\right)$ and $p_0 = N_h \exp\left(\frac{E_{\text{CBM}} - E_{\text{C}}}{k_B T}\right)$, where $T$ is the temperature, $k_B$ is the Boltzmann constant, $E_{\text{CBM}}$ is the Fermi level for $G = 0$, $E_{\text{CBM}}$ is the CBM level, $E_{\text{CBM}}$ is the CBM level, and $N_e$ and $N_h$ are the effective density of states for valence band and conduction band edges, respectively.

The SRH recombination coefficient $A$ can be calculated following

$$A = \frac{N_e c_{\text{SRH}} (n_0 + p_0 + \Delta n(0))}{c_0 (n_0 + n_1 + \Delta n(0) + c_{\text{Auger}} (p_0 + p_1 + \Delta n(0))},$$

where $N_e$ is the density of the recombination-center defects, $c_e$ and $c_p$ are the electron capture coefficient and hole capture coefficient of the defect level, respectively, and $n_1 = N_e \exp\left(\frac{E_{\text{CBM}} - E_{\text{C}}}{k_B T}\right)$ and $p_1 = N_h \exp\left(\frac{E_{\text{CBM}} - E_{\text{C}}}{k_B T}\right)$, where $E_{\text{C}}$ is the energy of the defect level.

The Auger recombination coefficient $C$ can be calculated following

$$C = \left[\gamma_n n_0 + \gamma_p p_0 (n_0 + p_0 + \Delta n(0))(p_0 + p_1 + \Delta n(0))\right] \frac{\Delta n(t)}{U_{\text{Auger}}(t)}.$$ (15)

In which $\gamma_n$ and $\gamma_p$ are the e-e-h Auger recombination coefficient (one electron at the conduction band is excited to a higher-level state when one electron–hole pair recombines) and the h-e-e Auger recombination coefficient (one hole at the valence band is promoted to a lower-level state when one electron–hole pair recombines).

According to equations (11) to (15), we can notice that the recombination rate $U(t)$ (including $U_{\text{SRH}}(t), U_{\text{band-band}}(t)$ and $U_{\text{Auger}}(t)$) depends nonlinearly on $\Delta n(t)$, so $\tau$ (the carrier lifetime) is not a constant with respect to $\Delta n(t)$. Therefore, in principle, the decay of $\Delta n(t)$ does not follow the simple function in equation (4). When $\tau$ was defined as the carrier lifetime (the time when the density decays to 1/e of the original value $\Delta n(0)$ after the generation stops) and was calculated by fitting the decay of $\Delta n(t)$ to the function $\exp(-t/\tau)$, we imposed implicitly that $\tau$ is a constant and $U(t)$ depends linearly on $\Delta n(t)$ in the following recombination processes after the generation stops. Of course, the fitting cannot be good if $\tau$ has a strong dependence on $\Delta n(t)$. Therefore, when we discuss the carrier lifetime $\tau$, we must pay attention to this dependence and note that $\tau$ can be approximated as a constant only for a small range of $t$ in which $\Delta n(t)$ does not change significantly. In such a small range of $t$, $\Delta n(t)$ can be approximated as $\Delta n(0)$, which is the value at the moment when the generation stops. With $\Delta n(0)$, the recombination coefficients $A$ and $C$ can be determined, then the corresponding lifetime can be calculated as

$$\tau_{\text{SRH}} = \frac{n_0 (n_0 + n_1 + \Delta n(0)) + c_{\text{Auger}} (p_0 + p_1 + \Delta n(0))}{N_e c_{\text{SRH}} (n_0 + p_0 + \Delta n(0))},$$

$$\tau_{\text{band-band}} = \frac{1}{B \times [n_0 + p_0 + \Delta n(0)]},$$

$$\tau_{\text{Auger}} = \frac{1}{\left[\gamma_n n_0 + \gamma_p p_0 (n_0 + p_0 + \Delta n(0))(p_0 + p_1 + \Delta n(0))\right] \Delta n(0)}.$$ (18)

In devices operating under continuous-light illumination, the density of non-equilibrium carriers should reach a steady state after the generation and recombination are counterbalanced, then the steady-state density should be considered as $\Delta n(0)$ when calculating the carrier lifetime according to equations (16), (17) and (18), as shown in Fig. 2a for the steady-state density in CH$_3$NH$_3$PbI$_3$ under light illumination.

As shown in Fig. 1a, defect-assisted SRH non-radiative recombination occurs between one carrier (electron or hole) and one defect, so the dependence of $U_{\text{SRH}}$ on $\Delta n$ (abbreviation of $\Delta n(0)$) is approximately first order in equation (11). The band-to-band recombination occurs between two carriers (one electron and one hole), so the dependence of $U_{\text{band-band}}$ on $\Delta n$ is approximately second order in equation (12). Auger recombination occurs between three carriers (two electrons and one hole, or one electron and two holes), so the dependence of $U_{\text{Auger}}$ on $\Delta n$ is approximately third order in equation (13). Therefore, when the density $\Delta n$ of photo-excited non-equilibrium carriers is small, $U_{\text{SRH}}$ and $U_{\text{band-band}}$ are much larger than $U_{\text{Auger}}$ and so $\tau_{\text{SRH}}$, $\tau_{\text{band-band}}$ determine $\tau$, but when $\Delta n$ is large, $U_{\text{SRH}}$, and $U_{\text{band-band}}$ are much smaller than $U_{\text{Auger}}$, so $\tau_{\text{Auger}}$ determines $\tau$ (ref. 19).

To calculate the total effective lifetime $\tau$ of non-equilibrium carriers in real semiconductor samples with a certain defect density $N_d$, equilibrium carrier density $n_0$, $p_0$, and non-equilibrium carrier density $\Delta n(0)$, $\tau_{\text{SRH}}, \tau_{\text{band-band}}$ and $\tau_{\text{Auger}}$ should all be calculated and combined. A procedure is plotted in Fig. 1b to show the flow of the calculations, which include the following:

(1) For a given type of defect with energy level $E_d$, calculate its electron and hole capture coefficients, $c_e$ and $c_p$. These can be calculated from the NAMD simulation or by using non-radiative multi-phonon (NMP) theory$^{19, 20, 21}$, as discussed
in the section ‘Calculation of defect-assisted SRH recombination lifetime’. With $E_c$, $c_{\text{e}, \Gamma_{\text{I}}}$ and $c_{\text{e}, \Gamma_{\text{T}1}}$ can be calculated for any given $N_c$, $n_c$, ($p_0$) and $\Delta r(0)$.

(2) Calculate the band-to-band recombination coefficient $B$. Details of the methods for this can be found in the section ‘Calculation of band-to-band recombination lifetime’. With $B$, $\tau_{\text{band-band}}$ can be calculated for any given $n_c$ ($p_0$) and $\Delta r(0)$.

(3) Calculate the Auger recombination coefficients $\gamma_n$ and $\gamma_p$. The methods for this are discussed in the section ‘Calculation of Auger recombination lifetime’. With $\gamma_n$ and $\gamma_p$, $\tau_{\text{Auger}}$ can be calculated for any given $n_c$ ($p_0$) and $\Delta r(0)$.

After the calculation of $\tau_{\text{Auger}}$, $\tau_{\text{band-band}}$ and $\tau_{\text{Auger}}$ the effective lifetime $\tau$ can be derived according to equation (16) and the value can be compared directly with the experimentally measured lifetime in real samples with given $N_c$, $n_c$ and $\Delta r(0)$.

Calculation of band-to-band recombination lifetime. As shown in Fig. 1a, there are two types of band-to-band recombination: radiative and non-radiative. For band-to-band radiative recombination, the energy of non-equilibrium carriers is converted into the energy of an emitted photon. For band-to-band non-radiative recombination, the energy of non-equilibrium carriers is converted into the energy of the vibration energy of ions through electron–phonon coupling. The recombination rate and lifetime are contributed by two parts, as described by the equation (16):

$$U_{\text{band-band}}(t) = \frac{1}{\tau_{\text{band-band}}(t)} + \frac{1}{\tau_{\text{non-rad}}(t)}$$

Where $\tau_{\text{band-band}}(t)$ and $\tau_{\text{non-rad}}(t)$ are the lifetimes for electrons and holes, respectively.

Using the DFT and plane-wave pseudopotential methods as implemented in the Vienna ab initio package (VASP) code. Starting from the optimized geometry of a 192-atom supercell at 0 K, the system is heated to 300 K using the velocity rescaling method for 2 ps. Then, 4- ps BOMD trajectories with a 1-fs timestep are generated in the microcanonical ensemble. All the geometries are used to calculate the non-adiabatic couplings (NAC) at the direct-bandgap $\Gamma$ point using PYPHAID. To simulate the charge carrier recombination processes, the 4-ps non-adiabatic Hamiltonians are iterated multiple times. Four hundred initial conditions are chosen randomly from the 4-ps trajectory to sample the canonical distribution of the atomic coordinates. After that, 2,000 realizations are performed to sample the real-time population. To simulate the band-to-band non-radiative recombination in the defect-free supercell, the electron is excited directly from the CBM. The system starts to enter the exponential function $\Delta t(0) \propto \exp(-t/\tau)$ (it can be approximated by a linear function $\Delta t(0) \propto 1 - t/\tau$) in a short timescale, which gives a lifetime $\tau$.

Band-to-band non-radiative recombination $\tau_{\text{non-rad}}$. In recent NAMD simulations of band-to-band recombination, carrier excitation is modeled through a change in the occupation numbers of the valence and conduction bands of the defect-free supercell. Recombination towards the ground state is then simulated by NAMD, and the band-to-band recombination lifetime $\tau$ is calculated by fitting the decay of the electron population using $\Delta n(t) \propto \exp(-t/\tau)$. These simulations are in the microcanonical (NVE) ensemble, so the electronic energy decrease is converted into vibrational (kinetic) energy through electron–phonon coupling, which obeys the energy conservation rule. Emission of light is not considered, so the NAMD-simulated band-to-band recombination is non-radiative and the associated lifetime is, in fact, $\tau_{\text{band-band}}$. This was also pointed out by Kim and Walsh.

Here we give an introduction to the calculation procedure for calculating $\tau_{\text{non-rad}}$ in the defect-free semiconductor (assuming CH3NH3PbI3, as an example) supercell. The supercell is divided into the energy of the supercell by the PYPHAID code for the first simulation (PYXHAID) code. We use the decoherence-induced surface hopping (DISH) approach to account for decoherence effect, which is required in the simulation of the recombination process across significant energy gaps. Before the NAMD simulation using PYXHAID, we first perform Born–Oppenheimer molecular dynamics (BOMD) and electronic structure calculations using the DFT and plane-wave pseudopotential methods as implemented in the Vienna ab initio package (VASP) code. Starting from the optimized geometry of a 192-atom supercell at 0 K, the system is heated to 300 K using the velocity rescaling method for 2 ps. Then, 4- ps BOMD trajectories with a 1-fs timestep are generated in the microcanonical ensemble. All the geometries are used to calculate the non-adiabatic couplings (NAC) at the direct-bandgap $\Gamma$ point using PYPHAID. To simulate the charge carrier recombination processes, the 4-ps non-adiabatic Hamiltonians are iterated multiple times. Four hundred initial conditions are chosen randomly from the 4-ps trajectory to sample the canonical distribution of the atomic coordinates. After that, 2,000 realizations are performed to sample the real-time population. To simulate the band-to-band non-radiative recombination in the defect-free supercell, the electron is excited directly from the CBM. The system starts to enter the exponential function $\Delta t(0) \propto \exp(-t/\tau)$ (it can be approximated by a linear function $\Delta t(0) \propto 1 - t/\tau$) in a short timescale, which gives a lifetime $\tau$.

According to the analysis, the short lifetime of 1.5 ns derived from NAMD simulations should be seriously underestimated and very short, ~1 ns, as demonstrated in Supplementary Fig. 1c for CH3NH3PbI3. The short lifetime from NAMD simulations is not the effective $\tau_{\text{non-rad}}$ for the total effective recombination rate in real samples, so it is meaningless to directly compare the value to the experimental lifetime.

To derive the supercell-size-independent recombination coefficient $B_{\text{non-rad}}$, we adopt equation (17) to derive the supercell-size-independent recombination coefficient $B_{\text{non-rad}}$ from the raw $\tau_{\text{non-rad}}$ derived from the NAMD supercell simulations. Equation (17) is changed to

$$P_{\text{non-rad}} = \frac{1}{\tau_{\text{non-rad}}(t)}$$

The densities of the equilibrium carriers, $n_c$ and $p_0$, should be small in the defect-free (dopant-free) supercell and can be neglected compared to $\Delta n(0)$, because the bandgap is above 1.5 eV and the thermal excitation of carriers can be neglected. As a result, $\tau_{\text{non-rad}}(t) \propto \tau_{\text{non-rad}}(0)$ and $\Delta n(0)$ can be as long as 10 ns, as demonstrated in Supplementary Fig. 1d. Based on the analysis, the short lifetime of 1.5 ns derived from NAMD simulations is not the effective $\tau_{\text{non-rad}}$ or the total effective $r$ in real samples, so it is meaningless to directly compare the value to the experimental lifetime.

A suitable way to predict the effective $\tau_{\text{non-rad}}$ is by adopting equation (17) to derive the supercell-size-independent recombination coefficient $B_{\text{non-rad}}$ from the raw $\tau_{\text{non-rad}}$ derived from the NAMD supercell simulations. Equation (17) is changed to

$$P_{\text{non-rad}} = \frac{1}{\tau_{\text{non-rad}}(t)}$$

The densities of the equilibrium carriers, $n_c$ and $p_0$, should be small in the defect-free (dopant-free) supercell and can be neglected compared to $\Delta n(0)$, because the bandgap is above 1.5 eV and the thermal excitation of carriers can be neglected. As a result, $\tau_{\text{non-rad}}(t) \propto \tau_{\text{non-rad}}(0)$ and $\Delta n(0)$ can be as long as 10 ns, as demonstrated in Supplementary Fig. 1d. Based on the analysis, the short lifetime of 1.5 ns derived from NAMD simulations is not the effective $\tau_{\text{non-rad}}$ or the total effective $r$ in real samples, so it is meaningless to directly compare the value to the experimental lifetime.

Calculation of defect-assisted SRH recombination lifetime. According to equation (16), $\tau_{\text{Auger}}$ can be calculated if the electron capture coefficient $c_e$ and hole capture coefficient $c_h$ of the defect level are known. $c_e$ and $c_h$ can be calculated using the NAMD simulation or NMP theory. The difference is that in the NAMD calculation procedure for $c_e$ and $c_h$ is similar to that for $B_{\text{non-rad}}$. The difference is that there is a recombination-center defect in the supercell
that produces a deep level in the bandgap, so the many-electron state basis sets of the NAMD simulation include the VBM, CBM and defect-level states. After an electron is excited from the VBM level to the CBM level, there are different transitions including those from the CBM to the defect level (the excited state to the intermediate defect-trap state in the many-electron state picture), the defect level to the VBM (the defect-trap state to the ground state) and from the CBM to the VBM (the excited state to the ground state). Their transition rates can be determined from the short-time DASH-based NAMD simulations, then the rates can be used to solve the defect-assisted coupled kinetic equations of electron–hole recombination in a nanosecond timescale. The calculated time evolution of the defect-trap state population can then be used to calculate the electron trapping time \( t_e \) and hole trapping time \( t_h \) of the defect level by exponential fitting, as described by Shi and colleagues.

Equation (16) and Supplementary Section 2 show the inversely proportional dependence of \( \tau_{\text{el}} \) (similarly for electron and hole trapping times \( t_e \) and \( t_h \)) on \( N_t \), so the derived lifetime from the NAMD simulations with one defect in a several-hundred-atom supercell (\( N_t \approx 10^4 \text{cm}^{-3} \)) should not be interpreted as the effective \( \tau_{\text{el}} \) (similarly for \( t_e \) and \( t_h \)) either. The lifetime fitted from the small-supercell NAMD simulations should be converted to the \( N_t \)-independent electron capture coefficient \( \gamma_e \) and hole capture coefficient \( \gamma_h \) of the defect according to equation (16), then the effective \( \tau_{\text{el}} \) in real samples with reasonable \( N_t \) can be calculated using the same equation. Because \( n_p, n_e, n_e \), and \( \gamma_e \) in equation (16) are all small for deep-level defects, but \( \Delta n(t) \) is very large in the NAMD simulation supercell, \( \tau_e = \frac{\lambda_e}{\gamma_e} \) for electron capture and \( \tau_h = \frac{\lambda_h}{\gamma_h} \) for hole capture. For example, Shi and colleagues performed NAMD simulations of the charge-trapping processes in CH3NH3PbBr3 containing the D1+ center. The obtained decay time, \( \tau_e = 1.9 \text{ ns} \) for CBM-to-defect trapping and \( \tau_h = 4.7 \text{ ns} \) for VBM-to-defect trapping. With the defect density (1.8 \( \times \text{10}^{13} \text{ cm}^{-3} \)) in their NAMD simulation, we can extract \( \lambda_e = 2.9 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1} \) and \( \lambda_h = 1.2 \times 10^{-11} \text{ cm}^3 \text{ s}^{-1} \). The values are low compared to the normal carrier capture coefficients of defects in semiconductors (10^-7 cm^3 s^-1). Assuming that \( N_t \) in real semiconductors is 10^13 cm^-3 and \( \Delta n(t) = 10^{10} \text{ cm}^{-3} \), \( \tau_{\text{el}} \approx 3 \times 10^4 \text{ s} \) according to equation (16). Therefore, according to the NAMD simulation, such a defect should not cause serious limit to the lifetime of photo-excited carriers in MAPbBr3, when the densities of the defect and photo-excited carriers are at a medium level.

As well as using the NAMD simulation, \( \gamma_e \) and \( \gamma_h \) can also be calculated using NTP theory based on the static coupling approximation derived from Fermi’s golden rule:\ref{14-15}

\[
\gamma_i = \langle f | H_{\text{e-ph}} | i \rangle^2 = \frac{2 \pi}{\hbar} \sum_{\text{phon}} \left| \langle f | \phi_{\text{phon}} \rangle \right|^2 \sum_{\text{phon}} \left| \langle i | \phi_{\text{phon}} \rangle \right|^2
\]

where \( V_i \) is the supercell volume, \( f \) is the Sommerfeld factor representing the Coulomb interaction between the charged defect and free carriers, \( \hbar \) is the reduced Planck constant, \( g_i \) is the degeneracy factor, \( \left| \langle f | \phi_{\text{phon}} \rangle \right|^2 \) is the electron–phonon coupling matrix element along phonon mode \( k \), \( \phi_{\text{phon}} \) is the Boltzmann occupation factor of the initial vibrational state of phonon state \( k \), \( \phi_{\text{phon}} \) is the initial and final vibrational wavefunctions, and \( E_k \) and \( E_{\text{phon}} \) are their eigenvalues. \( Q_0 \) refers to the geometry where the Hamiltonian \( H \) is expanded. By summing these quantities over all phonon modes using an infinite-integral scheme, one can obtain the carrier capture coefficient. Alternatively, adopting a single-phonon approximation simplifies the formula, and one can numerically calculate the integrals instead. A series of algorithms and codes have been developed for such calculations during the past decade.\ref{12,16-17}

When calculating coefficients \( \gamma_e \) and \( \gamma_h \) using either NAMD simulation or NTP theory, the convergence of the coefficients with respect to the supercell size should be tested, because the electron–phonon coupling effect for defects with different electronic states and phonon modes (localized or delocalized) can be quite different, and may not be accurately described by small supercells.\ref{18}

**Calculation of Auger recombination lifetime**

Equation (18) shows that \( \tau_{\text{Auger}} \) can be calculated when the e-e-h recombination coefficient \( \gamma_{\text{e-e-h}} \) and h-h-e coefficient \( \gamma_{\text{h-h-e}} \) are known. For direct Auger recombination, \( \gamma_{\text{e-e-h}} \) (similarly for \( \gamma_{\text{h-h-e}} \) can be calculated based on time-dependent perturbation theory using Fermi’s golden rule:\ref{19}

\[
\gamma_{\text{e-e-h}} = \frac{2 \pi}{\hbar} \sum_{ij} \sum_{\text{phon}} f_i (1 - f_i) (1 - f_j) |M_{ij\text{phon}}|^2 (E_i + E_j - E_k - E_{\text{phon}})
\]

where \( i \) and \( j \) indicate the single-particle states of electrons, and \( k \) of holes, \( f_i, f_j, f_k \) and \( f_h \) are the Fermi–Dirac occupation factors of these states depending on the quasi-Fermi levels for electrons and holes, which can be calculated in the same way as described in the calculation methods for \( B_{\text{Auger}} \). \( M_{ij\text{phon}} \) is the screened Auger matrix element, which can be calculated from the single-particle wavefunctions

\[
\psi_m = \psi_e |W| \psi_e \psi_h |W| \psi_h
\]

where \( \psi_e |W| \psi_e \) is the direct Coulomb term, and \( \psi_h |W| \psi_h \) is the exchange term.

An open-source code that implements this calculation method of the Auger recombination coefficients using the brute-force algorithm is available.\ref{19}

The calculated \( \gamma_e \) and \( \gamma_h \) are 2.7 \( \times \text{10}^{-10} \) and 4.6 \( \times \text{10}^{-10} \text{ cm}^{-3} \text{ s}^{-1} \) for CH3NH3PbI3 (ref. 23), and is close to our calculated value in Supplementary Section 3. The band-to-band radiative recombination coefficient \( B_{\text{rad}} \) was derived based on the NAMD simulation of CH3NH3PbI3 containing the DY and colleagues.\ref{22} The Auger recombination coefficients are \( \gamma_{\text{e-e-h}} = 2.7 \times 10^{-10} \text{ cm}^{-3} \text{ s}^{-1} \) and \( \gamma_{\text{h-h-e}} = 4.6 \times 10^{-10} \text{ cm}^{-3} \text{ s}^{-1} \). All these quantities can be calculated using the methods shown above and standard DFT calculations for any crystalline semiconductors.

With \( G \) and \( \Delta n(t) \) as a function of \( t \) can be simulated, as shown in Fig. 2a. At \( t = 0 \), \( \Delta n(t) = 0 \), then it starts to increase under the light illumination. As \( t \) increases, the system finally reaches the steady state and \( \Delta n(t) \) plateaus. As shown in Fig. 2a, \( \Delta n(t) \) reaches a steady value of 2.7 \( \times \text{10}^{-10} \) cm^3 s^-1 at \( t = 10 \) s when \( N_t = 10^3 \text{ cm}^{-3} \), whereas it reaches a steady value of 3.4 \( \times \text{10}^{-5} \) cm^3 s^-1 at \( t = 5 \) s when \( N_t = 10^8 \text{ cm}^{-3} \). Therefore, the steady density of non-equilibrium carriers and the time taken to reach the steady state are sensitive to the defect density. A higher defect density facilitates SRH recombination and thus decreases the time to reach the steady state.

In Fig. 2b we simulated the steady density of non-equilibrium carriers for various \( G \), corresponding to different illumination intensities. When \( G \) is below 10^8 cm^-3 s^-1, the steady-state density increases almost linearly as \( G \) increases. The generation rate under sunlight illumination is shown by the dashed line, and the steady density of non-equilibrium carriers (\( N_t \approx 10^8 \text{ cm}^{-3} \) for \( \gamma_{\text{e-e-h}} \)) reaches a steady value of 3.4 \( \times \text{10}^{-5} \) cm^3 s^-1, much lower than the imposed density of non-equilibrium carriers (\( \approx 10^{-4} \text{ cm}^{-3} \)) in small NAMD simulation supercells.
The steady density of non-equilibrium carriers under sunlight illumination should be taken as \( \Delta n(0) \) when calculating the lifetime of non-equilibrium carriers in CH\(_3\)NH\(_2\)PbI\(_3\)-based thin-film solar cells. Because the steady-state density changes with G, the lifetime should also change with G. When \( \Delta n(0) \), \( N \), \( n_F \), \( c_F \), \( n_0 \), \( n_F > n_0 \), \( R_{\text{band-band}} \), and \( R_{\text{Auger}} \) are known, \( \tau_{\text{rad}} \), \( \tau_{\text{band-band}} \), \( \tau_{\text{Auger}} \), and \( \tau_{\text{SRH}} \) can be calculated directly, following the procedure in Fig. 1b. Figure 2d-f show how G influences the calculated \( \tau_{\text{band-band}} \), \( \tau_{\text{Auger}} \), and \( \tau_{\text{SRH}} \) of CH\(_3\)NH\(_2\)PbI\(_3\). In Supplementary Section 4, we further demonstrate the calculation of effective carrier lifetimes in GaAs and CdTe with different carrier generation rates and different defect densities.

With the recombination rates of different mechanisms in equation (31), we can also calculate the internal PLQY:

\[
\eta = \frac{\tau_{\text{band-band}}}{\tau_{\text{band-band}} + \tau_{\text{Auger}} + \tau_{\text{SRH}}} = \frac{1}{1 + a \times (\Delta n(0) + \gamma \times \Delta n(0))^2},
\]

because only band-to-band radiative recombination is a radiative process, whereas the SRH, band-to-band non-radiative and Auger recombinations are non-radiative.

The calculated PLQY results are discussed in Supplementary Section 3.

**Data availability**

The raw data for the first-principles calculations have been deposited in the Zenodo repository\(^{24}\), including molecular dynamics calculations, non-adiabatic coupling calculations and momentum matrix elements calculations. Source data are provided with this Paper. Those data are generated by the code developed for this study.

**Code availability**

The effective carrier lifetime (ECL) code is published in the Code Ocean repository\(^{25}\).

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**Author contributions**

S.C. designed the research. S.W. and M.H. performed the calculations. All authors analysed the data, discussed the results and co-wrote the manuscript.

**Competing interests**

The authors declare no competing interests.

**Additional information**

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Extended Data Fig. 1 | Calculated non-equilibrium carrier density and carrier lifetime in GaAs under light illumination. a, The calculated density of non-equilibrium carriers in GaAs with varied carrier generation rate $G$. b–d, Effective carrier lifetimes for the SRH, band-to-band radiative and non-radiative, and Auger recombination in GaAs with varied $G$. The samples with different density $N_t$ of recombination-center defects are considered, (b) $N_t=10^8$, (c) $N_t=10^{13}$ and (d) $N_t=10^{15}$ cm$^{-3}$. The dashed line shows the lifetime calculated directly from the NAMD simulation of the band-to-band non-radiative recombination.
Extended Data Fig. 2 | Calculated non-equilibrium carrier density and carrier lifetime in CdTe under light illumination. a, The calculated density of non-equilibrium carriers in CdTe with varied carrier generation rate $G$. b–d, Effective carrier lifetimes for the SRH, band-to-band radiative and non-radiative, and Auger recombination in CdTe with varied $G$. The samples with different density $N_t$ of recombination-center defects are considered, (b) $N_t = 10^8$, (c) $N_t = 10^{13}$ and (d) $N_t = 10^{15}$ cm$^{-3}$. The dashed line shows the lifetime calculated directly from the NAMD simulation of the band-to-band non-radiative recombination.