Anomalous free-electron radiation beyond the conventional formation time

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Free-electron radiation is a fundamental photon emission process that is induced by fast-moving electrons interacting with optical media. Historically, it has been understood that, just like any other photon emission process, free-electron radiation must be constrained within a finite time interval known as the “formation time”, whose concept is applicable to both Cherenkov radiation and transition radiation, the two basic mechanisms describing radiation from a bulk medium and from an interface, respectively. Here we reveal an alternative mechanism of free-electron radiation far beyond the previously defined formation time. It occurs when a fast electron crosses the interface between vacuum and a plasmonic medium supporting bulk plasmons. While emitted continuously from the crossing point on the interface — thus consistent with the features of transition radiation — the anomalous radiation beyond the conventional formation time is supported by a long tail of bulk plasmons following the electron’s trajectory deep into the plasmonic medium. Such a plasmonic tail
mixes surface and bulk effects, and provides a sustained channel for electron-interface interaction. These results also settle the historical debate in Ferrell radiation, regarding whether it is a surface or bulk effect, from transition radiation or plasmonic oscillation.

The interaction between fast-moving electrons and optical media causes free-electron radiation. A famous example is Cherenkov radiation [1-6], in which photons are emitted from the bulk of a medium when the electron’s speed exceeds the speed of light in the medium. Another type of free-electron radiation, known as transition radiation [7-12], refers to photon emission from an interface—when an electron crosses an interface between different media, the electron will always emit photons, at any speed. As an alternative photon emission mechanism apart from atomic spontaneous emission and stimulated emission, free-electron radiation plays a significant role in many practical applications, ranging from high-energy particle detectors, free-electron lasers, electron microscopies, medical imaging, security scanning, to astronomy and cosmology [13-19].

In any type of radiation, it takes time for photons to be emitted, and the time interval is called the formation time. In the context of free-electron radiation, the concept of formation time in a bulk medium was firstly proposed by Ter-Mikaelian in Landau’s seminar in 1952 [20,21], and then further developed by Landau himself [22,23], with its physical effects experimentally demonstrated in 1990s [24-26]. Later, Ginzburg extended the formation time concept into photon emission from an interface, namely transition radiation [8,9,27]. As defined by Ginzburg, photons in transition radiation are emitted within one formation time [9]. This concept has already provided valuable guidance for practical applications. For example, the influence of formation time should be avoided in the design of transition radiation detectors [10], which are widely used in the identification of high-energy particles.

Here we reveal an alternative mechanism of free-electron radiation far beyond the previously defined formation time. It can occur when a fast-moving electron crosses the interface between free space and a plasmonic medium supporting bulk plasmons, such as metals at the
plasma frequency (Fig. 1). This radiation is supported by a long tail of bulk plasmons following the
electron’s trajectory deep into the plasmonic medium (see Supplementary Movie S1-S4 for a
demonstration). Such a plasmonic tail mixes the bulk and surface effects, making the radiation
neither a pure bulk effect as in Cherenkov radiation, nor a pure surface effect as in transition
radiation. A striking feature for this anomalous radiation, as we will demonstrate later, is its long
duration far beyond the formation time historically defined for free-electron radiation.

It is interesting to note that such a geometry of electrons bombarding plasmonic media has
been long studied in the field of plasmonics since 1950s. In fact, the existence of surface plasmons
was firstly confirmed with electrons bombarding a metal film [28]. However, the complex electron-
photon-plasmon interaction has left a few issues in the history that have not been fully resolved. A
typical example is the Ferrell radiation [29,30] (i.e., the enhanced radiation at the plasma frequency
of the bulk medium), which could be ascribed to a surface effect with pure transition radiation, or
a bulk effect induced by plasmonic oscillation, or both, but so far there is no decisive
conclusion [31-38] (see supplementary section S5 for a historical survey). With our revealed
mechanism of anomalous free-electron radiation beyond the conventional formation time, it
becomes feasible to settle this historical debate here.

We consider in Fig. 1 that a fast electron moves with a velocity $\vec{v} = \hat{z}v$ and penetrates the
interface separating region 1 and region 2, where their relative permittivity is $\varepsilon_{1r} = 1$ (free space)
and $\varepsilon_{2r}$ (either a dielectric or a plasmonic medium), respectively, $v = 0.4c$, and $c$ is the speed of
light in free space. By imposing the constraint of $\varepsilon_{2r} < (c/v)^2 = 6.25$, we can exclude the
possibility of Cherenkov radiation, since the electron’s speed falls below the Cherenkov threshold.
Consequently, the only possible radiation, according to conventional theories of classical
electrodynamics, should be transition radiation, as studied by Ginzburg and many other
colleagues [9,12,27,39]. Since we are interested in the backward radiation propagating almost
parallel to the electron’s trajectory, the conventional formation time of transition radiation, as
defined by Ginzburg, is \( t_f(\omega) = \frac{2\pi}{\omega|1 + v_1\sqrt{\varepsilon_1r}|} + \frac{2\pi}{\omega|1 - v_2\sqrt{\varepsilon_2r}|} \) [8, 9]; see Methods for details. To facilitate
the discussion, \( t_{f_0} = t_f(\omega_p) \) is chosen as a reference to normalize the horizontal coordinate of time in Fig. 2. Accordingly, the length of formation zone, also known as the formation length or coherence length [8, 20, 21, 40], is \( L_{f_0} = vt_{f_0} \).

Now we consider a plasmonic medium in region 2 with \( \varepsilon_2r = \varepsilon_{Drude}(\omega) \). To capture the role of losses, we employ a Drude-like formula to describe the relative permittivity of plasmonic media, namely \( \varepsilon_{Drude}(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega/\tau} \), where the plasma frequency is set to be \( \omega_p = 13.9 \) petahertz, and \( \tau \) is the relaxation time; note that the nonlocal response of plasmonic media has a minor influence on the radiation revealed here; see Figs. S7-S8. As a comparison, we also consider the control situation when region 2 is a regular dielectric with \( \varepsilon_2r = 2 \).

Figure 2a-f shows the temporal evolution of the backward-radiation field at a point \( \vec{r}_{\text{far}} \) far away from the interface but close to the electron’s trajectory, where the angle between \( \vec{r}_{\text{far}} \) and \( -\vec{z} \) is \( \theta_{\text{far}} \approx 7^\circ \). The major part of emitted photons at \( \theta_{\text{far}} \) is formed within \( t_{f_0} \), as shown in Fig. 2a & d. If the fast electron is within the conventional formation zone of transition radiation, according to Ginzburg’s analysis, the electron can directly interact with the interface. Then the significant process of photon emission within the conventional formation time in Fig. 2a & d can be treated as a surface effect. Beyond \( t_{f_0} \), if region 2 is a plasmonic medium with \( \varepsilon_2r = \varepsilon_{Drude}(\omega) \), there are sizable radiation fields that oscillate periodically far beyond \( t_{f_0} \) in Fig. 2a-b, where the relaxation time \( \tau = \tau_0 \) is used and \( \tau_0 = 700/\omega_p \). In contrast, if region 2 is a dielectric with \( \varepsilon_2r = 2 \), the radiation field beyond \( t_{f_0} \) is negligible as shown in Fig. 2d-e.

Moreover, the radiation fields beyond the conventional formation time \( t_{f_0} \) in Fig. 2b have their frequency close to the plasma frequency; see the frequency-domain analysis in Fig. 2c. Figure 2c shows that after the electron crosses the interface, the radiation peak around the plasma
frequency is mainly contributed by the emission process of photons beyond the conventional formation time $t_f(\omega \approx \omega_p) \approx t_{f_0}$. This way, we denote the radiation field beyond $t_{f_0}$ as the anomalous free-electron radiation, while denoting the radiation field within $t_{f_0}$ as the conventional free-electron radiation (or transition radiation).

Figure 3a-d shows that the revealed anomalous radiation is caused by the interaction between the interface and a long tail of excited bulk plasmons (both the transverse and longitudinal electromagnetic waves are considered for bulk plasmons; see supplementary section S7). The reason is that the tail of excited bulk plasmons in the plasmonic medium not only follows the electron’s trajectory but also can attach to (and thus interact with) the interface for a long time (Fig. 3a). In other words, even when the fast electron is far beyond the formation zone of transition radiation, the electron will continue to interact with the interface at the same crossing point; this way, the radiation process is unfinished beyond the conventional formation time, and the fast electron will continue to emit photons from the interface (Fig. 3a). Since the excitation of bulk plasmons is a bulk response of the plasmonic medium, it is reasonable to treat the formation of anomalous radiation as a mixture of surface-bulk effect, instead of solely a surface effect. Besides, since the surface plasmons at the surface of plasmonic media exist far below the plasma frequency $\omega_p$ (i.e., $\omega < \omega_p/\sqrt{2}$) but the excited bulk plasmons have their frequency close to $\omega_p$, the surface plasmons cannot be excited by the bulk plasmons but can be excited only by the direct electron-interface interaction within the conventional formation time [27]; see more discussion about Fig. 3a-d in Methods.

Due to the interface-bulk effect, we shall re-define the formation time for this anomalous free-electron radiation. This formation time, labelled as $T_{e-\text{interface}}$, describes the electron-interface interaction time and is here defined as the time taken for the tail of bulk plasmons to leave the interface, which can be treated simply as the ratio between the full length of the tail (created by a fast electron moving inside a homogeneous plasmonic medium) and the electron’s velocity.
Apparently, this formation time $T_{e\text{-interface}}$ is highly dependent on the loss of plasmonic media. For example, $T_{e\text{-interface}} > 100t_0$ if $\tau > 0.1\tau_0$, as shown in Fig. 3e. The large value of $T_{e\text{-interface}}$ directly indicates that the anomalous free-electron radiation can occur far beyond the conventional formation time of transition radiation. This anomalous free-electron radiation is distinct from the conventional free-electron radiation trapped by resonators (inside which the out-coupling process of the already-formed photons can last for a relatively-long time without the existence of electron-resonator interactions, if the quality factor of resonators is high); see Methods.

To further understand the anomalous radiation, we explore it in the frequency domain in Fig. 4. Figure 4a-b shows the angular spectral energy density $U_1(\omega, \theta)$ of backward radiation. From Fig. 4a, there is a radiation peak near the plasma frequency if region 2 is the plasmonic medium. This radiation peak is historically known as the Ferrell radiation [11,29-38,41], which has caused a long debate about its physical origin, regarding whether it is a surface or bulk effect, from transition radiation or plasmonic oscillation (see supplementary section S5 for a detailed historical survey). Nowadays, most literatures have ascribed Ferrell radiation to radiative surface plasmons or the so-called Ferrell mode [30,36,38,41], but a complete picture is still lacking to fully settle the debate.

We find that the introduction of the anomalous free-electron radiation can address this issue. Firstly, there are two types of plasmonic oscillations simultaneously, bulk plasmons in the bulk and radiative surface plasmons on the interface. While bulk plasmons are excited as the plasmonic tail deep into the plasmonic medium, the tail also touches the interface, exciting radiative surface plasmons. Secondly, while it is the radiative surface plasmons that directly emit photons from the interface (but that alone cannot continuously emit photons far beyond the conventional formation time due to their large decay rate), it is the bulk plasmons that provide the energy, and the bulk plasmons in turn draw energy from the fast-moving electron. Along the way, the electromagnetic field extends coherently from the fast-moving electron to the interface. In other words, it takes a
long distance, and thus a long time interval, to “peel off” the electromagnetic field from the electron, eventually forming photons. Thirdly, the Ferrell radiation is contributed by both transition radiation and plasmonic oscillation. These two contributions are impossible to be distinguished in the frequency domain but can be separated in the time domain. The transition radiation, as described by Ginzburg, occurs only within one conventional formation time. The significant radiation beyond the conventional formation time comes from the plasmonic oscillation, as we have analyzed in Fig. 2.

In conclusion, we have revealed the emergence of anomalous free-electron radiation beyond the conventional formation time by investigating the penetration of a fast-moving electron through the interface of a plasmonic medium. This anomalous radiation is closely related to a long tail of bulk plasmons, which follows the electron’s trajectory deep into the plasmonic medium and can attach to and thus interact with the interface for a very long time. Correspondingly, this tail provides a unique route to mix the surface and bulk effects, significantly extend the electron-interface interaction, and then create light emission far beyond the conventional formation time. Therefore, the revealed anomalous free-electron radiation is intrinsically a mixture of surface-bulk effect, distinct from Cherenkov radiation as purely a bulk effect or transition radiation as purely a surface effect. Our finding also provides a new perspective for Ferrell radiation and settles its historical debate regarding its physical origin. The anomalous free-electron radiation may further trigger many open questions, concerning, for example, the observation of the long tail of bulk plasmons or the anomalous radiation beyond the conventional formation time, the possibility to largely enhance particle-light-matter interactions by exploiting bulk plasmons in other types of free-electron radiation such as Smith-Purcell radiation and synchrotron radiation, and the design of advanced light sources based on the anomalous radiation.
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Author contributions
All authors contributed extensively to this work. X.L., X.S., and B.Z. initiated the idea; F.T., X.L., and X.S. performed the calculation; I.K., B.Z., and H.C. contributed extensively to the data analysis and explanation of detailed results; F.T., X.L., and B.Z. wrote the manuscript with the input from the other authors; X.L., B.Z., and I.K. supervised the project.

Competing interests
The authors declare no competing interests.

Additional information
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Methods
Derivation of free-electron radiation from an interface
When a fast electron crosses an interface, free-electron radiation will happen, and it can be analytically calculated in the framework of Maxwell’s equations. Such a derivation is given in section S1 in the supporting information. Based on the analytical results of the charge field and radiation field in the frequency domain, the temporal dynamics of the charge field and radiation field can be further calculated numerically.

Conventional formation time and formation zone
The definition of conventional formation time and formation zone for transition radiation, according to Ginzburg’s work [8,9], is introduced in Section S2. The conventional formation time depends on the frequency and the radiation angle.

More frequency-domain study on the backward free-electron radiation
With the knowledge of the radiation fields in free space or region 1, the angular spectral energy density and the energy spectral density are derived in section S3. In addition to Fig. 4, Fig. S1 shows the angular spectral energy density for the temporal study of the backward free-electron radiation in Fig. 2a-b; Figure S2 shows more analysis on the angular spectral energy density with \( \varepsilon_{2r} = \varepsilon_{\text{Drude}}(\omega) \) in Fig. 4a by using various values of \( \tau \).
More temporal study on the anomalous free-electron radiation
More discussion on the temporal study of the anomalous free-electron radiation is given in section S4. For example, Fig. S3 provides more analysis of the anomalous free-electron radiation in Fig. 2a-c by setting $\tau = 0.1\tau_0$ & $0.01\tau_0$. Figure S4 shows the field distribution when a fast electron moves in a homogeneous material.

Ferrell radiation
The interesting history of Ferrell radiation, which is fraught with controversies at the very beginning, is briefly introduced in section S5.

Dependence of the long tail of bulk plasmons on the electron’s velocity
The long tail of bulk plasmons inside the plasmonic medium under different electron’s velocities is investigated in section S6. The appearance of this tail is irrelevant to the electron’s velocity, but its shape will change with the electron’s velocity, as shown in Fig. S5. Moreover, the influence of the electron’s velocity on the anomalous free-electron radiation is discussed in Fig. S6. The intensity of anomalous free-electron radiation increases with the electron’s velocity.

Influence of the nonlocal response of plasmonic media and the longitudinal electromagnetic waves on the anomalous free-electron radiation
As the longitudinal wave may appear when the permittivity of the plasmonic medium is close to zero, the influence of the longitudinal waves on the anomalous free-electron radiation is studied in section S7. To tackle this issue, the plasmonic medium is modelled by the hydrodynamic model in the derivation, which takes the nonlocal response of the plasmonic medium into consideration. Figure S7 shows the influence of the nonlocal response of the plasmonic medium and the longitudinal wave on the distribution of total fields when the electron perpendicularly crosses an interface between free space and a plasmonic medium. Figure S8 studies the influence of the nonlocal response and the longitudinal wave on the anomalous free-electron radiation, both in the frequency and time domains. We find the consideration of the nonlocal response and the longitudinal wave has a trivial influence on the formation of anomalous free-electron radiation revealed here.

More analysis of Fig. 3a-d: excited surface plasmons and photons below the plasma frequency
Discussion on the excited surface plasmons below the plasma frequency: When a swift electron perpendicularly crosses the interface between vacuum and a plasmonic medium, the field distribution of $E_y$ in the time domain, including the charge field and radiation field, is plotted in Fig. 3a-d. If the interface between regions 1 and 2 supports surface plasmons, the swift electron will excite both photons and surface plasmons. For the surface plasmons at the interface of the vacuum-plasmonic medium, they exist only at the frequency below $\omega_p/\sqrt{2}$. This way, the long tail of bulk plasmons (having a frequency close to $\omega_p$) will not lead to the excitation of surface plasmons. As such, we mainly focus on the discussion of the emission process of photons, instead of the emission process of surface plasmons, in the main text. For the surface plasmons at the interface of the vacuum-plasmonic medium, their group velocity, along with their propagation length, decreases with the frequency. This way, when the electron is far away from the interface, there are only the excited surface plasmons, which have their frequency close to $\omega_p/\sqrt{2}$, in the region close to the electron’s trajectory if $\tau$ is large (e.g., $\tau = 0.1\tau_0$ in Fig. 3a); If $\tau$ is small (e.g., $\tau \leq 0.1\tau_0$ in Fig. 3b-c), there are no apparent excited surface plasmons in the region near the electron’s trajectory, due to their large propagation loss.
Discussion on the emitted photons below the plasma frequency: When the swift electron is far away from the interface, apart from the field of excited surface plasmons, there are some radiation fields of photons in the region of free space close to the electron’s trajectory in Fig. 3a-d. These radiation
fields of photons are partially contributed by the emitted photons with a low frequency, because the formation length of photons in free space increases when the frequency decreases and can reach tremendous values if the frequency is low enough. In other words, for the emitted low-frequency photons, the swift electron in Fig. 3a-d (even though far away from the interface) can still be within their formation zone; namely, the emission process of low-frequency photons may be not finished in Fig. 3a-d. Moreover, note that Fig. 3a-d plots $E_z$ instead of $D_z$, and $D_z$ is continuous on the interface, i.e., $\varepsilon_{1f}(\omega)E_{1z}(\omega) = \varepsilon_{2f}(\omega)E_{2z}(\omega)$. Then it is sensible that the radiation field $E_z$ of photons in region 1 (free space) is much larger than that in region 2, since at low frequency we have $|\varepsilon_{2f}(\omega)| > |\varepsilon_{1f}(\omega)|$ (no matter $\varepsilon_{2f} = 2$ or $\varepsilon_{2f} = \varepsilon_{\text{Drude}}(\omega)$).

Based on the above analysis, when the electron is far away from the interface, it is reasonable to argue that the total field in Fig. 3a-d is dominated by the charge field, and the radiation field (including both photons and surface plasmons) is trivial since the major radiation field propagates far away from the interface.

**Difference between the anomalous free-electron radiation and the conventional free-electron radiation from resonators with a high-quality factor**

When a fast electron interacts with resonators with a high-quality factor, the excited photons inside the resonator may be out-coupled into the surrounding environment (e.g., free space) with a very low decay rate. This might lead to a relatively-long total time for the whole radiation process. Due to the low decay rate of the resonator, the whole radiation process can be well separated into two processes, namely the photon-formation process and the follow-up photon out-coupling process. To be specific, the photons are mainly created during the finite-time interaction between the fast electron and the resonator (dependent on the finite size of resonators), and this photon-formation process for the resonator case still obeys the conventional formation time-zone effect. On the other hand, during the follow-up continuing photon out-coupling process, the particle-resonator interaction disappears, and the relatively-long out-coupling process is purely caused by the ultra-low decay rate of the already-formed photons inside the resonator with a high-quality factor. Therefore, the whole free-electron radiation process from resonators with a high-quality factor is intrinsically different from the anomalous free-electron radiation beyond the conventional formation time revealed here. For the anomalous free-electron radiation in our case, the whole radiation process has the electron-interface interaction supported via the long tail of bulk plasmons inside the plasmonic medium. Moreover, the radiative surface plasmons in our case once created generally has a large decay rate, indicating that the related photon out-coupling process will be relatively short, and the radiative surface plasmons alone in our case cannot lead to the whole radiation process far beyond the conventional formation time.

**Data availability**

The data that support the plots within this paper and other finding of this study are available from the corresponding author upon reasonable request.

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Fig. 1. Schematic of the anomalous free-electron radiation beyond the conventional formation time. A fast-moving electron with velocity \( \vec{v} = z \vec{v} \) impinges on an interface and induces radiation, where \( v = 0.4c \) is used. The interface separates regions 1 (namely air) and region 2 (a dielectric or a plasmonic medium), whose relative permittivity is \( \varepsilon_{1r} \) and \( \varepsilon_{2r} \), respectively. The anomalous radiation occurs due to the formation of a long tail of bulk plasmons when the electron moves inside the plasmonic medium, whose optical response is described by a Drude-like formula, namely

\[
\varepsilon_{\text{Drude}}(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega/\tau}.
\]
Fig. 2. Representation in the time domain of the anomalous free-electron radiation beyond the conventional formation time. The structural setup is the same as Fig. 1. (a) Temporal evolution of the backward-radiation field $E_p(\vec{r}_{\text{far}}, t)$ at a point $\vec{r}_{\text{far}}$ far away from the interface but close to the electron trajectory when region 2 is filled by a plasmonic medium with $\varepsilon_{2r} = \varepsilon_{\text{Drude}}(\omega)$. (b) An enlarged plot of the radiation field beyond the conventional formation time (highlighted by a purple dashed square in (a)). (c) Frequency-domain analysis of the radiation, namely the Fourier transform.
transform of (a) and (b), which are carried out in different ranges of time as specified in each panel.

(d-f) Conventional free-electron radiation when region 2 is filled by a regular dielectric with $\varepsilon_2 = 2$. All analysis in (d-f) is the same as (a-c). Here and below, we set $t = 0$ as the moment when the electron enters the formation zone in the air, $t_r = t - \frac{r_{\text{far}}}{c}$ as the retarded time, $r_{\text{far}} = |\vec{r}_{\text{far}}|$, $t_{f0} = t_f(\omega = \omega_p)$, and $t_f(\omega)$ is the conventional formation time defined for transition radiation.
Fig. 3. Spacetime-domain illustration of the underlying physics for the anomalous free-electron radiation. The anomalous radiation is caused by the indirect electron-interface interaction, which is mediated via a long tail of bulk plasmons and can exist far beyond the conventional formation time. (a-d) Spatial distribution of the electric field $E_x(\vec{r}, t)$; see also in Movies S1-S4. (e) Electron-interface interaction time $T_e$ as a function of the relaxation time $\tau$. $T_e$ can far exceed $t_{f0}$, if $\tau$ increases or if $|\varepsilon_{\text{Drude}}(\omega_p, \tau)\rangle$ (instead of merely $\text{Im}(\varepsilon_{\text{Drude}}(\omega_p, \tau))$) decreases down to a minor value. The shaded region at the interface in (b) represents the formation length of transition radiation, $L_{f0}$. Besides, $\lambda_p = 2\pi c/\omega_p$. 
**Fig. 4. Frequency-domain analysis of the anomalous free-electron radiation.** The structural setup is the same as Fig. 1. (a, b) Angular spectral energy density $U_1(\omega, \theta)$ of the backward radiation. Region 2 is filled by a plasmonic medium with $\varepsilon_{2r} = \varepsilon_{\text{Drude}}(\omega)$ in (a) and a regular dielectric with $\varepsilon_{2r} = 2$ in (b).