Charge equilibration is a rapid process during the early stage of heavy-ion collisions. A basic mechanism of charge equilibration is presented in terms of the extension of single-particle motion from one nucleus to the other, from which the upper energy-limit of the bombarding energy is introduced for significant charge equilibration at the early stage of the collision. The formula for this limit is presented, and is compared to various experimental data. It is examined also by comparison to three-dimensional time-dependent density functional calculations. The suppression of charge equilibration, which appears in collisions at the energies beyond the upper energy-limit, gives rise to remarkable effects on the synthesis of exotic nuclei with extreme proton-neutron asymmetry.

Because this spreading occurs as a consequence of unblocked single-particle motion, the process can be very fast; a particle travels into the other side within $\sim 10^{-22}$ sec at a quarter of light velocity, which roughly corresponds to the Fermi energy of normal nuclear matter. On the other hand, this propagation can be prohibited by the Pauli principle, while the neutron (or proton) excess weakens this effect. Another important factor is the difference of velocities of the two nuclei. Because this spreading needs a certain time, it does not lead to charge equilibration in the initial stage if the relative velocity of the colliding nuclei is too large. In the charge equilibration, protons and neutrons from both nuclei, particularly those near the Fermi levels, are mixed within a certain time after the touching.

We can introduce an ansatz that, in order for the fast charge equilibration to occur, the relative velocity $v_r$ of the two nuclei at the collision must be below the velocities corresponding to the proton or neutron Fermi momenta of both nuclei. By denoting the minimum of these four velocities as $v_F^{\text{min}}$, the present effect can be summarized by the statement that the upper limit of the bombarding energy for charge equilibration is determined by $v_r = v_F^{\text{min}}$. The upper limit of the energy in the laboratory frame for charge equilibration is expressed as the sum of the kinetic energy for velocity $v_F^{\text{min}}$ and the Coulomb energy at touching:

$$ E_{\text{CE,lab}}/A \text{[MeV]} = \frac{\hbar^2(3\pi^2 \rho_{\text{min}})^{2/3}}{2m} + \frac{e^2 Z_1 Z_2}{4\pi\epsilon_0 r_0} A_1 + A_2 \left( A_1^{1/3} + A_2^{1/3} \right)^{1/3}, \quad (1) $$

$$ \rho_{\text{min}} = \min_i \left( \rho_{ni}, \rho_{zi} \right) = \min_i \left( \frac{N_i \left( 4\pi \epsilon_0 A_i^{1/3} \right)^{-1}}{(1-3\epsilon_0)(1+\delta)}, \frac{Z_i \left( 4\pi \epsilon_0 A_i^{1/3} \right)^{-1}}{(1-3\epsilon_0)(1-\delta)} \right), \quad (2) $$

where $m$, $e$, $\epsilon_0$, and $r_0$ are the nucleon mass, the charge
By collecting TDHF events, we sort them with weights of geometric cross section, and elastic cases. These events are summed over impact parameters values of impact parameter being incremented by 0.25 cm. Many TDHF events were obtained with different perform three-dimensional TDHF calculations systematic. skins.

A $\varepsilon$ formula is used to calculate the proton or neutron density of the two nuclei using the radius parameter (1.2 fm), respectively. Here we express in Eq. (2), $i_1$ and $\bar{i}_2$ (functions of $A_i$) are introduced based on the droplet model to take into account the effect of neutron and proton skins.

In order to confirm the validity of this picture, we perform three-dimensional TDHF calculations systematically. Many TDHF events were obtained with different values of impact parameter being incremented by 0.25 fm. These events are summed over impact parameters with weights of geometric cross section, and elastic cases are discarded. By collecting TDHF events, we sort them in terms of $N/Z$ ratio with $N/Z$ being discretized into bins of width 0.05. Figure 1 shows the yield distribution of final fragments for $^{208}$Pb + $^{132}$Sn reaction. Going from low to high $E_{cm}$ (total kinetic energy in the center-of-mass frame) of the collision, the peak energy of the yield distribution as a function of $N/Z$ is almost constant at the beginning, and is shifted later with the lowering of peak height. A clear decrease of the yield of charge-equilibrated fragments for $E_{cm}/A \geq 7.0$ MeV is noticed. On the other hand, very neutron-rich nuclei with $N/Z \sim 2.0$ simultaneously start to be produced. By taking $E_1$ as the energy at which the peak height starts to lower (6.0 MeV in this case), and $E_2$ as the energy at which the yield of the equilibrated $N/Z$ becomes about 50 % (7.0 MeV in this case), the upper energy-limit is defined in TDHF as $E_{CE} = (E_1 + E_2)/2$ (6.5 MeV in this case). The uncertainty from the energy bin value is $(E_2 - E_1)/2$ (0.5 MeV in this case). Such TDHF results are summarized in the third and forth columns of Table I. TDHF calculations with two different parameter-sets result in the same upper energy-limit. Note that a large value is obtained for $^{24}$Mg + $^{24}$O, implying that a simple mass dependence (small values for reactions between light nuclei, and vice versa) cannot explain this, while the mass asymmetry plays a certain role.

Let us have a comparison between the present upper-limit and the TDHF results. Figure 2 depicts how the upper limit given by Eq. (1) changes as functions of $N/Z$ and $A_1/A_2$ (or $A_2/A_1$). The upper energy-limit comes down significantly low for higher mass asymmetry, while it depends only weakly on the total mass. Although charge equilibration can compete with Coulomb excitation particularly in collisions involving nuclei with large $Z$, no evidence of a major change due to large $Z$ is found in the results of Eq. (1) or those of TDHF calculations. However, non-negligible decrease of the upper energy-limit due to the proton-neutron asymmetry is noticed. The corresponding values obtained by the formula in Eq. (1) is shown in the fifth column of Table I. For reference, the values obtained by the simple Fermi gas model ($k_F = 1.36$ fm$^{-1}$ for both protons and neutrons) are shown in its sixth column. By comparing the results of Eq. (1) with the TDHF results shown in the third and forth columns, the agreement is remarkable, including a high value in the last row. Comparison between the results of Eq. (1) (blue circles) and the TDHF results (blue bars) is also made in Fig. 2. Consequently,
where equilibration based on Eq. (1) in the center-of-mass frame, FIG. 2: 

\[ \frac{N}{Z} \] dependence of the upper-limit of charge equilibration based on Eq. (1) in the center-of-mass frame, where \( A_1 > A_2 \) is assumed without loss of generality. Values with different total masses \( A = 100, 200, 300, 400, \) and 500 are plotted for each \( A_1/A_2 \), which correspond to black lines from bottom to top in each group (values are too crowded to distinguish the total mass difference, for the cases of \( A_1/A_2 = 10 \) and 100), and lines are drawn to guide eyes. Each TDHF calculation is shown as a blue bar, where the central points correspond to the value obtained by Eq. (1) for each reaction, and Roman numbers distinguish reactions shown in Table I.

the upper-limit of charge equilibration depends largely on the Fermi energy, and the proton-neutron asymmetry can contribute to the shift of the upper-limit, where the dependence of the upper-limit on the mass-asymmetry is remarkable.

Let us move on to the comparison to the experiments. It can be seen that existing experimental data agrees with the present upper-limit formula. For instance, the following experiments show charge equilibration: \(^{40}\)Ar + \(^{58}\)Ni at \( E_{lab}/A = 7.0 \text{ MeV} \) [11] and \(^{56}\)Fe + \(^{105}\)Ho (\(^{209}\)Bi) at \( E_{lab}/A = 8.3 \text{ MeV} \) [12]. The following experiment shows the disappearance of charge equilibration: \(^{112}\)Sn + \(^{124}\)Sn at \( E_{lab}/A = 50 \text{ MeV} \) [13, 14]. Recently, experiments: \(^{124,112}\)Sn + \(^{124,112}\)Sn at \( E_{lab}/A = 35 \) and 50 MeV respectively was performed at Michigan State University [16]. One remarkable result here is that the final fragments are not so close to charge equilibrium even in lower energy collisions with \( E_{lab}/A = 35 \text{ MeV} \). The disappearance of charge equilibration for this experiment has not been explained. Because the upper limit of charge equilibration is calculated to be \( E_{lab}/A = 27.6 \text{ MeV} \) from Eq. (1), this question can be understood now.

The suppression of charge equilibration contribute naturally to the production of exotic fragment; more exotic nuclei far from the equilibrated \( N/Z \) ratio are to be synthesized above the present upper-limit. Because the bombarding energies of the 3rd generation RI-beam facilities are sufficiently high to exceed the upper-limit, the present upper-limit ensures more production of further exotic isotopes by the latest and the future RI-beam facilities. In fact, in the experiment of Ref. [15], the yield of exotic fragments was increased simply by putting the beam energy higher than the present upper-limit. It is also of much interest to explore the novel possibility of the synthesis of exotic nuclei by collisions including only \( \beta \)-stable nuclei. Such possibilities have not attracted much attention, but may emerge with various feasibilities of the production of exotic species, if the experiment is set for energies beyond the present upper-limit. As an example, Fig. 3 shows the real-time dynamics of \(^{208}\)Pb + \(^{40}\)Ca around the energy where the production of exotic nuclei starts; the upper energy limit is 3.66 MeV (See Table I, fusion appears at \( E_{cm}/A = 3.0 \text{ MeV} \) (upper panels), and break-up is seen at 4.0 MeV (lower panels). \( \beta \)-unstable fragments are emitted only for the bombarding energy higher than the upper-limit; for instance, a small fragment in the lower-right panel of Fig. 3 is \(^{62}\)Mn (numbers of nucleon are rounded to be integer), for which the stable isotope is \(^{55}\)Mn.

We now point out some interesting details of charge-equilibration dynamics for collisions below the present upper energy limit. We shall first investigate it with a focus on the iv-GDR in a collision between light nuclei. Figure 4 (a) shows the time-evolution of charge distribution for \(^{24}\)O + \(^{24}\)Mg. We see the appearance of the iv-GDR and the oscillation of well-localized charge (4.5, 6.0 \times 10^{-22}\text{ sec}). The charge equilibration is synchronized with the iv-GDR. This is, however, seen only for
collisions between light nuclei. Similar results are obtained in [2,8]. Next, we move on to collisions involving a heavier nucleus; e.g. $^{24}\text{Mg} + ^{208}\text{Pb}$, as shown in Fig. 4 (b). Charge-equilibration dynamics is not similar to the motion of the iv-GDR in this case, because no significant oscillations of the localized charge appear. Instead a radially layered structure of the composite nucleus is formed (7.5 x 10^{-22} sec), in which a relatively neutron-rich core appears in the center, a proton-rich layer surrounds it, and a neutron-rich skin is in the surface. This seems to be due to the Coulomb repulsion, hence the radial distribution of charge is formed. Here one can find analogous situation of isovector monopole excitation. It means that the radial flow plays a prominent role in such low energy collisions. Similar layered structures are obtained in the TDHF calculations (with both Skyrme parameter-sets) listed in Table I except for $^{24}\text{Mg} + ^{24}\text{O}$. Consequently, charge equilibration, which is dependent on the mass, has been clarified to be related not only with the isovector giant dipole excitations, but also with the isovector giant monopole excitations.

Finally, let us comment on the diffusion-type mechanism towards charge equilibrium. The mechanism of charge equilibration discussed in this Letter is valid at lower energies (lower than the upper energy-limit). At much higher energies, collisions between nucleons and the diffusion contribute mostly to charge equilibration (for example, see [17,18]). As the diffusion is a slow process, it is unlikely to attain the charge equilibrium within the initial stage. Energy-dependent equilibration mechanisms of a similar kind have been reported in condensed matter physics (see [12-21]), where the equilibration is fast for lower energies (temperatures), and quite slow for higher energies. Such a similarity between completely different physical systems is interesting.

In this Letter the mechanism of charge equilibration, in which nucleons with the Fermi velocity play a primary role, has been presented, and its validity is examined by comparison to virtually all existing relevant experimental data. This concept has been further analyzed in terms of a three-dimensional time-dependent density function formalism. The upper energy-limit of charge equilibration has a crucial impact on the nuclear synthesis of exotic nuclei. Properties presented in this Letter will give a sound motivation for the production of further exotic isotopes by the latest and the future RI-beam facilities.

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[1] H. Freiesleben and J.V. Kratz, Phys. Rep. 106 (1984) 1.
[2] M. Berlanger, A. Gobbi, F. Hanepepe, U Lynen, C. Ngô and A. Olmi, Z. Phys. A 291 (1979) 133.
[3] E.S. Hernandez, W.D. Myers, J. Randrup and B. Remaud, Nucl. Phys. A 361 (1981) 483.
[4] P. Bonche and N. Ngô, Phys. Lett. B 105 (1981) 17.
[5] E. Suraud, M. Pi, and P. Schuck, Nucl. Phys. A 492 (1989) 294.
[6] V. Baran, D.M. Brink, M. Colonna, and M. Di Toro, Phys. Rev. Lett. 87 (2007) 182501.
[7] C. Simenel, Ph. Chomaz, and G. de France, Phys. Rev. Lett. (2001) 2971.
[8] C. Simenel, Ph. Chomaz, and G. de France, Phys. Rev. C 76 (2007) 024609.
[9] W.D. Myers and W.J. Swiatecki, Ann. Phys. 55 (1969) 395.
[10] W.D. Myers, Phys. Lett. 30B (1969) 451.
[11] B. Gatty, D. Guerreau, M. Lefort, X. Tarrago and J. Galin, Nucl. Phys. A 253 (1975) 511.
[12] H. Breuer et. al., Phys. Rev. Lett. 43 (1979) 191.
[13] P. M. Milazzo et. al., Phys. Rev. C66 (2002) 021601.
[14] W.P. Tan et. al., Phys. Rev. C64 (2001) 051901.
[15] M. Mocko et. al., Phys. Rev. C76 (2007) 014609.
[16] M.B. Tsang et. al., Private communication.
[17] L. Shi and P. Danielewicz, Phys. Rev. C 68 (2003) 064604.
[18] M.B. Tsang et. al., Phys. Rev. Lett. 102 (2009) 122701.
[19] L.D. Landau and Zh. Eksperim, Soviet Phys. - JETP 5 (1957) 101.
[20] W.R. Abel, A.C. Anderson, and J.C. Wheatley, Phys. Rev. Lett. 17 2 (1966) 74.
[21] A.J. Leggett, Rev. Mod. Phys. 47 No.2 (1975) 331.