Direct Experimental Evidence of Nonequilibrium Energy Sharing in Dissipative Collisions

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Primary and secondary masses of heavy reaction products have been deduced from kinematics and energy–time-of-flight measurements, respectively, for the direct and reverse collisions of ¹⁰⁰Mo with ¹²⁰Sn at 14.1*A* MeV. Direct experimental evidence of the correlation of energy sharing with net mass transfer and model-independent results on the evolution of the average excitation from equal-energy to equal-temperature partition are presented. [S0031-9007(97)02323-5]

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The determination of the microscopic mechanism of energy dissipation and energy partition between the reaction partners of a dissipative collision has been a controversial subject of debate in the past years [1-12] (for a review, see [13]). The excitation energy sharing presents an evolution with the inelasticity of the reaction: In quasielastic events the two reaction partners reseparate with almost equal excitation energies, but with increasing dissipation there is a trend towards equilibrium partition (i.e., excitation energy shared in proportion to the mass of the fragments). In most cases, however, such a condition seems not to be reached even for the largest dissipated energies [2,6,9,11]. These experimental findings can be explained by models [14] which describe the evolution of many macroscopic observables by means of stochastic exchanges of single nucleons between the interacting nuclei. More refined experiments [4,7,10] claimed that the excitation energy division is correlated with the net mass transfer, with an excess of excitation being deposited in the fragment which gains nucleons. Moreover, the strength of this experimental correlation seems to be largely independent of the degree of inelasticity [10] and this latter result seems difficult to understand within a stochastic nucleon exchange picture. The present Letter aims to obtain, in a model independent way, direct experimental information on this subject.

In a previous paper [15], we used the sequential fission to investigate the degree of equilibration between the two reaction partners at the end of the interaction. This was achieved with an asymmetric system (120 Sn + 100 Mo at 19.1*A* MeV) in which a given primary mass *A* corresponds to different net mass transfers for projectile- and targetlike fragments (PLF and TLF). The striking result was that the curves of fission probability P_{fiss} vs fissioning mass for PLF and TLF do not coincide: For a given *A*, P_{fiss} for the TLF (which gained mass) was significantly larger than for the PLF, even at large total kinetic energy losses (TKEL). However, sequential fission, while providing a tool of great sensitivity, did not allow us to determine which variable (among those relevant to fission, e.g., excitation energy, isospin, angular momentum, deformation) is mainly responsible for the observed effect. Moreover, fission events form a somewhat biased sample and may be not fully representative of all events of a dissipative collision within a specific TKEL bin.

In order to investigate whether similar nonequilibrium effects are present also in the two-body exit channel, we turned to a different tool, namely the light particle evaporation, which depends mainly on the excitation energy. This procedure was applied in the past to measurement of PLF from rather asymmetric systems studied in direct kinematics only [4,8,10] and required a detailed and not trivial comparison between the experimental results and evaporation calculations. To avoid relying on model calculations (which become increasingly uncertain with increasing excitation energy), we aimed at comparing not the data with a model, but directly two sets of experimental data.

With an asymmetric colliding system, one might compare the two event samples in which reaction products of a given mass *A* are PLF or TLF, this fact implying different "histories" (gained or lost nucleons). To overcome the severe experimental difficulties (like threshold effects, poor resolution, and critical dead layer corrections) which impede the measurement of the TLF with sufficient accuracy, we devised the alternative approach of measuring the secondary mass of the PLF only, however, studying the same asymmetric collision both in direct and reverse kinematics. This approach gives also the additional bonus that the efficiencies for the detection of the PLF, being quite similar for the two kinematics, practically do not affect the result of the comparison.

This Letter presents for the first time a direct experimental evidence (based not on comparison with evaporation models, but on experimental results only) indicating that the number of emitted nucleons depends on the net mass transfer experienced by the primary reaction products. This observation is strongly suggestive of a nonequilibrated sharing of the excitation energy between the two reaction partners. Beams from the Unilac accelerator of GSI-Darmstadt were used to study the asymmetric collision 100 Mo + 120 Sn at 14.1 *A* MeV, both in direct and reverse kinematics. The moderate asymmetry of the entrance channel was chosen in order to make sure that a common range of masses for PLF and TLF was available even at not too large TKEL The chosen isotopes, having almost the same N/Z ratio (1.38 and 1.40 for 100 Mo and 120 Sn, respectively), ensure that isospin equilibration plays a negligible role. The experiment was based on the measurement of both the primary (via the kinematic coincidence method, KCM) and secondary mass (via additional measurement of the kinetic energy) of the PLF.

Heavy ($A \ge 20$) products were detected in an array of 12 position-sensitive parallel-plate avalanche detectors (PPAD), covering about 75% of the forward hemisphere [16,17]. The FWHM resolutions of time of flight and position were 700 ps and 3.5 mm, respectively. From the measured velocity vectors, primary (preevaporative) quantities were deduced event by event with an improved version of the KCM [18]. The FWHM resolution on the primary mass values is of the order of 3%-4%. The background of incompletely measured events of higher multiplicity was estimated [18] and subtracted.

An array of 40 Si detectors of various sizes (from $1 \times 1 \text{ cm}^2$ at small polar angles, up to $5 \times 5 \text{ cm}^2$), of 300 μ m thickness, was mounted behind two of the forward PPAD, so as to cover a sizable part of the region below and around the grazing angle, where partly damped events are concentrated ($\theta_{\text{graz}}^{\text{lab}} \approx 10^\circ$ for the present collisions). Secondary masses A_{sec} of the PLF were obtained (with a FWHM resolution of about 5%–6%) event by event from the energy deposited in the Si detectors and the time-of-flight measured by the corresponding PPAD, using an iterative procedure which takes into account the pulse height defect in the semiconductors and the energy loss in the PPAD and in the dead layers.

For various windows of TKEL (corrected for the Q_{gg} between entrance and exit channel [17]), the experimental data were sampled in bins of reconstructed primary mass A of the PLF and the centroids of the corresponding distributions of evaporated mass $\Delta A = A - A_{sec}$ were determined. The full squares and full circles in Fig. 1 show ΔA as a function of the primary mass of the PLF in the direct and reverse reaction, respectively. For the two kinematic cases, one observes two distinct (but almost parallel) linear rises of ΔA with increasing primary mass A. The presented data refer to the region TKEL \leq 500 MeV, corresponding to partly damped events, where PLF can be safely distinguished from TLF due to the strongly anisotropic angular distributions [16].

Quantities like the primary mass A and TKEL are correlated to a certain extent with each other (being obtained from the same velocity vectors), as well as with the secondary mass A_{sec} (via time of flight). Moreover, the overall finite resolution (arising both from the smearing of the



FIG. 1. The full squares (circles) show the experimental average number of evaporated nucleons ΔA as a function of the primary mass A of the PLF in the direct (reverse) reaction, for windows of TKEL. ΔA is the difference between the primary mass and the centroid of the corresponding distribution of secondary masses. The open symbols show the experimental data after correction for the response of the setup, finite resolution effects, and distortions of the analysis.

particle evaporation process and from the detection procedure) can cause systematic distortions in determining the value of nonuniformly distributed variables (see, e.g., the comment about the angular distribution in [18] and the correction of the mass distribution in [19]). As analytic corrections may be worked out only in very simple cases, the experimental results were corrected via extensive Monte Carlo simulations, modeling the dissipative collision followed by an evaporative emission in agreement with the statistical code GEMINI [20] and incorporating as realistically as possible the response of the setup, finite resolution effects, and all known distortions of the analysis method. The open symbols in Fig. 1 show the experimental data after correction.

The most striking result resides in the different values of ΔA obtained, for a given A, in the direct and reverse collisions. It has to be noted that the differences (4– 6 amu) between the evaporated masses in the two cases are much larger than the applied corrections (1–2 amu at most). We want also to stress that the corrections to be applied to the experimental ΔA (and hence the corrected data points in Fig. 1), are within errors largely independent of physical hypothesis (e.g., on energy partition), as it was checked by repeating the Monte Carlo simulations with different physical models. From the comparison between the two sets of corrected experimental points of Fig. 1 one obtains information on the mechanism of excitation energy sharing. The striking difference in ΔA between the two kinematic cases can be viewed as a dependence of the excitation energy sharing on the net mass transfer. This behavior is put here into evidence without recourse to statistical model calculations (our use of Monte Carlo simulated data is limited to the correction for experimental systematic effects). We recall that none of the usual ways of modeling the excitation energy sharing—neither the equal-energy, nor the equal-temperature scenarios, nor any combination of the two—foresees the observed splitting of the correlation ΔA vs A into two well separated branches.

Just to clarify this point, let us focus the attention on the symmetric exit channel, in which the two primary fragments have the same mass number A = 110. If the dinuclear system at reseparation had lost memory of its history, the two excited reaction products should have deexcited by emission of the same average number of nucleons, irrespective of the size of the fluctuations in the internal degrees of freedom. Thus the observed difference in ΔA indicates a sizable deviation from equilibrium at the end of the interaction phase. Actually, due to the enhanced sensitivity of the particle evaporation process to the excitation energy of the emitter (with respect to other internal variables like isospin, angular momentum [2] or deformation), this experimental result is a proof of a sizable deviation from equilibrium in the excitation energy sharing. Neglecting preequilibrium emission and evaporation from the dinucleus during the interaction phase (which are small at these bombarding energies [5,16]), one can estimate the mean excitation energy ϵ removed per evaporated nucleon. Dividing the central value of the TKEL bin by the sum of the masses evaporated by the PLF with A = 110 in the direct and reverse reaction, one obtains, at all TKEL, values of ϵ (\approx 11–12 MeV) which are in good agreement with the 12–13 MeV predicted by GEMINI. Thus one roughly estimates that, of two nuclei of primary mass A = 110, the one obtained by a gain of ten nucleons (PLF in the direct reaction) should be about 50-60 MeV more excited than the one obtained by removal of ten nucleons (PLF in the reverse reaction).

More quantitatively, one can build the ratio

$$R = (\Delta A_{110}^l - \Delta A_{110}^h) / (\Delta A_{110}^l + \Delta A_{110}^h), \quad (1)$$

where ΔA_{110}^l (ΔA_{110}^h) is the total evaporated mass for nuclei with primary mass A = 110, originating from the entrance channel light (heavy) nucleus and measured as PLF in direct (reverse) kinematics. Figure 2(a) shows the so defined *R* as a function of TKEL. *R* is an estimate of the excitation-energy asymmetry $(E_{110}^{*l} - E_{110}^{*h})/(E_{110}^{*l} + E_{110}^{*h})$, being $E_{110}^{*l} = \epsilon \Delta A_{110}^l$ ($E_{110}^{*h} = \epsilon \Delta A_{110}^l$) the excitation energy of nuclei with A = 110, originating from the entrance channel light (heavy) nucleus.



FIG. 2. (a) Asymmetry in the total evaporated mass $R = (\Delta A_{110}^l - \Delta A_{110}^h)/(\Delta A_{110}^l + \Delta A_{110}^h)$ for nuclei of primary mass A = 110, originating from the entrance channel light and heavy nuclei, as a function of TKEL. R is an estimate of the excitation-energy asymmetry due to the net gain or loss of nucleons. (b) Asymmetry in the evaporated mass $C = (\Delta A_{120}^h - \Delta A_{100}^l)/(\Delta A_{120}^h + \Delta A_{100}^l)$ for nuclei with A = 100 and 120 originating from the entrance channel light and heavy nucleus, respectively, as a function of TKEL. C is an estimate of the excitation energy partition in absence of net mass transfer: C = 0 indicates equipartition of excitation (equal energy sharing); the dotted line shows the value expected for thermal equilibrium (equal temperature sharing).

In the present experiment also the average partition of excitation energy between the two reaction partners can be deduced — in a substantially model-independent way — from the number of nucleons emitted in case of no net mass transfer. Figure 2(b) shows the ratio

$$C = (\Delta A_{120}^h - \Delta A_{100}^l) / (\Delta A_{120}^h + \Delta A_{100}^l)$$
(2)

as a function of TKEL, where $\Delta A_{100}^l (\Delta A_{120}^h)$ is the total evaporated mass for nuclei of primary mass A = 100and 120 originating from the entrance channel light and heavy nucleus, respectively. *C* is an estimate of the excitation energy partition $(E_{120}^{*h} - E_{100}^{*l})/(E_{120}^{*h} + E_{100}^{*l})$, when assuming a common value ϵ for the average energy necessary to evaporate a single nucleon from the two reaction partners (we verified with GEMINI that indeed the actual values of ϵ for A = 100, 120 differ by no more than 1% to 3% when E^* ranges from 50 to 250 MeV).

Our data show that for the exit channel without net mass transfer the total excitation energy is initially almost equally shared between the fragments ($C \approx 0$ at small TKEL) and that the approach to the equilibrium partition [mass ratio, shown by the dotted line in Fig. 2(b)] is very slow with increasing TKEL. In spite of the limited range of *C* values spanned between these two extremes (less then 10%, due to the moderate mass asymmetry of the collision), we find that even at the highest explored TKEL

a partition consistent with full thermal equilibrium is not reached. This average behavior is in agreement with other experimental results and compatible, by itself, with model descriptions based on the exchange of independent nucleons during the contact phase.

A linear dependence of the energy partition on the net mass transfer had been proposed by Toke *et al.* [10]:

$$E_{\rm PLF}^{*}(A)/E_{\rm tot}^{*} = C_T + R_T(A - A_{\rm beam}),$$
 (3)

where the excitation energy of a PLF of mass A was given in terms of the TKEL-dependent parameters C_T (describing the partition in case of no mass transfer) and R_T (describing the rise of ΔA with A in Fig. 1).

Using the parameters defined in Eqs. (1) and (2), we can write Eq. (3) for the products deriving from the original light or heavy colliding nucleus:

$$\frac{E^{*\,l,h}(A)}{E_{\rm tot}^*} = \frac{1}{2} + \frac{C}{A_{\rm dif}} \left(A - \frac{A_{\rm tot}}{2} \right) + \frac{R}{A_{\rm dif}} \left(A - A_0^{l,h} \right),\tag{4}$$

where A_0^l (A_0^h) is the lighter (heavier) mass between A_{beam} and A_{target} in the entrance channel, $A_{\text{tot}} = A_0^l + A_0^h$ and $A_{\rm dif} = \tilde{A}_0^h - A_0^l$. Our notation has the advantage of making evident that there is a mass dependent term (containing C) which simply describes the dependence of excitation energy on mass (e.g., in case of thermal equilibrium, $C = A_{\rm dif}/A_{\rm tot}$ leading to a trivial proportionality to the mass of the nucleus). However, only the term containing R truly represents a net-mass-transfer dependent term and it is responsible for the splitting of the correlation into two distinct branches. The slope parameter R_T of Eq. (3) mixes the two terms as it comes out to be $R_T \equiv (C + R)/A_{\text{dif}}$. We want to stress that the experimental decomposition of R_T in the two contributions was possible only in the present experiment, due to the measurement of the PLF in the direct and reverse kinematics. Previous experiments, measuring the evaporated mass of PLF in one kinematic case only, could attempt such a decomposition only in a model-dependent way.

The observed correlation between "evaporated mass" and net mass transfer, which is here evidenced without need of model calculations, strongly suggests that there is no complete equilibrium between the two reaction partners. The persisting strength of such a correlation even at high TKEL is a strong challenge for a microscopic description. Within a nucleon exchange picture, some correlation might arise from a possible donor-acceptor intrinsic asymmetry in the excitation energy deposition caused by the exchange of a single nucleon. However, with increasing dissipation, the larger and larger number of independent nucleon exchanges should almost wash out the correlation. Toke et al. [10] emphasized the surprising constancy of the donor-acceptor intrinsic asymmetry η which they deduced from a reanalysis of the data of Ref. [4]. Following Ref. [10], which employed, as usual, the experimental mass variances σ_A^2 as an estimate of the total number of

exchanges, we obtain unreasonable values from our data (η comes out to be greater than unity and increases with increasing TKEL), also because of the very rapid increase of σ_A^2 with TKEL. In the spirit of Ref. [10], also this fact points to a failure in the present description of the nucleon transfer process at larger bombarding energies. Indeed, with increasing TKEL and bombarding energy, other effects might come into play, which cannot be described simply with the elementary process of single nucleon transfer across a window. For example, remaining in the framework of one-body dissipation picture, the rapid increase of σ_A^2 with TKEL could be reconciled with a smaller number of exchanges if a relevant contribution comes from the transfer of clusters of nucleons. Alternatively, a relevant role could be played by collective effects, such as formation and rupture of a neck during the collision. Both of these suggestions require complete and precise theoretical calculations.

In conclusion, the existence of a mechanism which correlates the evaporated mass—and hence the excitation energy sharing—with the net mass transfer has been evidenced in a model independent way. This experimental finding seems difficult to reconcile with existing models based on stochastic exchanges of singles nucleons and calls for a better theoretical understanding of the microscopic interaction mechanism of heavy nuclei.

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