high-Z intermediate nuclei would be of great value in this context.

The observed fission-fragment mass distributions show the well-known asymmetric mass split in the quasielastic region with a rapid transition to symmetric fission for larger energy losses. The Z dependence of the rms width of these distributions, plotted in Fig. 2(e), exhibits a considerable increase in the region of Z > 100 up to the heaviest elements, much larger than expected<sup>10</sup> for the slight increase in nuclear temperature, correlated with Z. A possibly related broadening in the mass distribution has also recently been observed in fusion-fission reactions of lighter elements with fission barriers reaching zero because of very high angular momenta.<sup>9</sup> This apparent lack of dependence on the entrance channel suggests that the phenomenon reflects inherent properties of the liquiddrop model, i.e., a correlation between a decreasing stability against mass asymmetry and a loss of stability in the fission degree of freedom for nuclei with a vanishing fission barrier. Alternatively again, a relatively fast process may be involved, in which the shape of the observed mass distributions is influenced by an incomplete thermalization of the asymmetry mode, strongly excited in the first step of the reaction.11

In summary, our investigations of the heaviest collision systems available have so far not given evidence for an instantaneous three-body breakup. Irrespective of the apparent two-step nature of the observed fission phenomena with relatively long scission-to-scission times of  $\ge 10^{-20}$  s, non-equilibrated systems may still be involved. Their clear identification has to be left to future experiments.

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## Evidence for a Soft Nuclear-Matter Equation of State

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The entropy of the fireball formed in central collisions of heavy nuclei at center-ofmass kinetic energies of a few hundred MeV per nucleon is estimated from the ratio of deuterons to protons at large transverse momentum. The observed paucity of deuterons suggests that strong attractive forces are present in hot, dense nuclear matter, or that degrees of freedom beyond the nucleon and pion may already be realized at an excitation energy of 100 MeV per baryon.

One of the principal motivations for accelerating heavy-ion beams to relativistic energies is the hope of producing and studying matter at baryon densities greater than are found in atomic nuclei. However, information about the properties of the dense matter thus created is obscured by the fact that the matter remains hot and dense only for a very short time,  $< 10^{-22}$  sec, and our observations are limited to the products emitted as it disassembles. We present arguments that the ratio  $R_{dp}$  of deuterons to protons is established during the early stages of the fireball's existence, and is little changed by the later processes of expansion and disassembly. We show that the ratio  $R_{dp}$  is proportional to the density of neutrons in phase space (momentum  $\times$  position), and thus measures the specific entropy of the fireball. Applying our arguments to recent measurements<sup>1</sup> of Ne + NaF and Ar + KCl at 100 to 200 MeV/nucleon c.m. kinetic energy, we find that a surprisingly large region of phase space is populated. This could be explained by strong attractive interactions in the hot dense matter, or by a proliferation of the degrees of freedom sharing the excitation energy.

It has been argued<sup>2</sup> that the time evolution of the fireball in central collisions goes through three stages: formation, explosion, and disintegration. In collisions of large nuclei, with relative velocity greater than the speed of sound, a fireball may be formed at high density. At high energies or for small nuclei, there may be partial interpenetration, in which case the initial density of the hot matter will be less than double normal density. During this stage of the reaction, most of the kinetic energy of the nuclei's relative motion is thought to be converted to thermal excitation of the dense matter. A great deal of entropy is created in this stage of the reaction.<sup>3</sup>

During the second stage of the reaction, repeated collisions lead to a hydrodynamic expansion of the matter. This expansion is largely reversible, so that little additional entropy is generated during the expansion.<sup>2,3</sup> The additional phase space due to the expansion of matter into a larger region of position space is compensated by the shrinking of the velocity distribution into a smaller region of momentum space as the expanding matter cools. During this stage of the reaction, entropy is produced by viscous forces and by thermal conduction. Heat conduction produces entropy at a rate proportional to the square of the thermal gradients; so it should not be very large. The viscous damping of the shear-velocity gradients also produces entropy, but this merely completes the initial thermalization process.

In the third stage of the reaction, the density of particles has become so small that they seldom collide. This stage lasts until the particles reach the detectors. During this stage, the matter is no longer described hydrodynamically because the distribution of momenta at any given position and time cannot be characterized by a mean velocity and a thermal fluctuation. Instead, strong correlations develop between position and momentum, which may be used to identify the particles in a time-of-flight telescope or magnetic spectrometer. However, Liouville's theorem quarantees that the particles' density in phase space remains constant in the absence of collisions.

Because of the reaction  $d+N \leftrightarrow p+n+N$ , where N is a spectator nucleon or cluster, deuterons will be constantly breaking up and reforming. If collisions are frequent enough, the deuterons will quickly reach an equilibrium concentration determined by detailed balancing<sup>4</sup>:

$$\exp(-\mu_d/T)d_d(\mathbf{\bar{R}}, \mathbf{\bar{P}}, \mathbf{S}_z) = \sum_{s_z} d_p(\mathbf{\bar{R}}, \mathbf{\bar{P}}/2, s_z)d_n(\mathbf{\bar{R}}, \mathbf{\bar{P}}/2, S_z - s_z)\exp[-(\mu_n + \mu_p)/T],$$
(1)

where  $d_i(\mathbf{R}, \mathbf{P}, S_z)$  is the six-dimensional phase-space density (Wigner mixed density) of species *i* at position  $\mathbf{R}$  with momentum  $\mathbf{P}$  and spin projection  $S_z$ ,  $\mu_i$  is its chemical potential, and *T* is the temperature of the equilibrium thermal distribution. Since the deuteron is a very weakly bound system we expect  $\mu_d - \mu_n - \mu_p \ll T$ . Summing over spins and momenta, we obtain for the ratio of deuterons to protons

$$R_{dp} = [(2S_d + 1)/(2S_p + 1)] \otimes \langle d_N \rangle,$$

where  $\langle d_N \rangle = \iint d^3p \, d^3r [d_N(\mathbf{\tilde{p}}, \mathbf{\tilde{r}})]^2 / \iint d^3p \, d^3r \, d_N(\mathbf{\tilde{p}}, \mathbf{\tilde{r}})$  is the average phase-space density of neutrons, and  $d_n = d_p = d_N$  is assumed.

The rate of approach to the equilibrium concentration is governed by the collision frequency, with a time constant equal  $to^4$ 

$$\tau = [n_N \langle \sigma(N + d \rightarrow n + p + N) v_{rel} \rangle]^{-1},$$

where  $n_N$  is the nucleonic density. Since the measured values of  $\sigma(N + d \rightarrow n + p + N)$  are similar to elastic nucleon cross sections, the equilibration of deuteron number will be just as rapid as the thermalization of the nucleonic momentum distributions. Thus  $R_{dp}$  will be in equilibrium through the hydrodynamic expansion.<sup>5</sup> In fact, since  $R_{dp}$  is just given by the mean density of nucleons in phase space  $\langle d_N \rangle$ , we can relate  $R_{dp}$  to the entropy per nucleon  $S_N$  carried by the nucleons

$$S_N = \frac{5}{2} - \ln(2^{2/3} \langle d_N \rangle) = 3.95 - \ln R_{db}.$$
(3)

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(2)

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Thus  $\langle d_N \rangle$  and  $R_{dp}$  will be approximately constant throughout the hydrodynamic-expansion phase.

Even after the collisions cease to maintain hydrodynamic local equilibrium, the number of deuterons does not change. If the remaining interactions are too weak to significantly alter the phase-space distribution  $d_N$ , then an alternative estimate of the number of deuterons is obtained by projecting the uncorrelated neutron and proton densities onto the deuteron wave function,<sup>6</sup>

$$d_{d}(\vec{\mathbf{R}}, \vec{\mathbf{P}}, \mathbf{S}_{z}) = \sum_{s_{z}} \iint d^{3}r \, d^{3}p \, d_{p}(\vec{\mathbf{R}} + \frac{1}{2}\vec{\mathbf{r}}, P + \frac{1}{2}\vec{p}, \mathbf{S}_{z} + \mathbf{S}_{z}) d_{n}(\vec{\mathbf{R}} - \frac{1}{2}\vec{\mathbf{r}}, \vec{\mathbf{P}} - \frac{1}{2}\vec{p}, \mathbf{S}_{z} - \mathbf{S}_{z}) d_{in}(\vec{\mathbf{r}}, \vec{p}, \mathbf{S}_{z}).$$
(4)

If  $d_N$  varies slowly over the range of the deuteron's intrinsic structure

$$d_{\mathrm{in}}(\mathbf{\vec{r}},\mathbf{\vec{p}}) = (2\pi)^{-3} \int d^3 \mathbf{r}' \,\psi_d(\mathbf{\vec{r}} + \frac{1}{2}\mathbf{\vec{r}}')\psi_d(\mathbf{\vec{r}} - \frac{1}{2}\mathbf{\vec{r}}') \exp(i\,\mathbf{\vec{p}}\cdot\mathbf{\vec{r}}'),$$

we recover the relations (1) and (2) [neglecting  $(\mu_d - \mu_n - \mu_p)/T$ ]. From Liouville's theorem,  $\langle d_N \rangle$  does not change after the collisions cease. We conclude that the ratio of deuterons to protons in the final state measures the entropy of the neutrons in the fireball, according to Eq. (3).

The functions  $d_i(\mathbf{\tilde{r}}, \mathbf{\tilde{p}})$  describe the distribution of particles in an individual collision. The observed distributions, on the other hand, are averages over collisions with different impact parameters and different reaction planes. Since relation (1) is nonlinear, it will not in general be satisfied if average quantities are substituted. To minimize this difficulty, we follow Ref. 2 and restrict our analysis to measurements of similarsized target and projectile at large transverse momentum. This kinematic regime is associated with central collisions of high multiplicity. The measured distributions at 90° c.m. are fitted with

a radially expanding Boltzmann distribution as in Ref. 2. The temperature and expansion velocity are determined from fits to the tails of pion and proton spectra; the ratio  $R_{dp}$  is then determined by the ratio of the normalizations. Sample fits are shown in Fig. 1. The resulting entropies are plotted in Fig. 2. The error bars are made large enough to include the value of  $R_{dp}$  from the total differential cross sections at  $90^{\circ}$  c.m., which include some peripherally produced protons and deuterons of small transverse momentum. We see that the entropy decreases for larger systems or more central collisions, which would be expected since the thermalization takes place at higher mean densities in these cases. The energy per baryon is taken as the beam energy minus the



FIG. 1. Spectra at 90° c.m. for reactions  $A_p + A_T \rightarrow d + X$ . Data are from Lemaire *et al.*, Ref. 1. Theoretical curves are exploding-fireball model from Siemens and Rasmussen, Ref. 2, with temperature and velocity fitted to proton and pion spectra, normalization fitted to measured points.



FIG. 2. Entropy per baryon as a function of excitation energy per baryon for various reactions, including one with measured associated multiplicities greater than 3. Theoretical curves are for soft nuclear-matter equation of state at compression ratios of 1, 2, and 4 [Eq. (5) of the text, with K = 200 MeV].

energy of observed pions (assuming isospin symmetry) at 90° c.m.; the error bar indicates the size of the pion subtraction. We see that the entropy increases with energy, as expected.

To judge the significance of the results, we show the predictions for entropy versus excitation energy for a very soft equation of state at various densities in Fig. 2. The energy per baryon is taken from a model of nucleons moving freely in a potential whose density dependence is chosen so that the speed of sound in cold matter is independent of density at T = 0:

$$E(n,S) = E(n_0,0) + \frac{1}{9}K[(n_0/n) - 1 + \ln(n/n_0)] + E_{FG}(n,S) - E_{FG}(n,0),$$
(5)

where *n* is the baryon number density,  $n_0$  is the density of normal matter with compressibility  $K_{\bullet}$ and  $E_{FG}(n,S)$  is the energy per baryon of a free Fermi gas [evaluated in the limit of low degeneracy, corresponding to Eq. (3). We see that much more entropy is created than would be expected from even such a soft equation of state for nucleons as this. Even the inclusion of real pions in the system does not give enough entropy. This is especially surprising in light of the fact that such diverse dynamical models as fluid dynamics<sup>7</sup> and intranuclear cascade<sup>8</sup> predict maximum compressions of three to four.

The interesting conclusion that seems to be forced upon us is that more entropy is generated during the collision than we would naively expect. It could be that there are strong attractive forces<sup>9</sup> present in the hot, dense nuclear medium, thus raising the entropy. It could also be that many more mesonic and baryonic particulate degrees of freedom are excited at this energy than suspected.<sup>10</sup> Or it could be that collective degrees of freedom, such as due to pion condensation,<sup>11</sup> are the culprits. Or perhaps the nucleons dissociate into quarks. It is impossible to distinguish among these possibilities from purely thermodynamic measurements, without involving detailed theoretical models.

In any event it is important to make measurements for excitation functions, and to use heavier nuclei to reduce any unwanted surface effects. Clearly it is also important to perform more detailed numerical calculations to further support the scenario described in this Letter. These could take the form of deuteron production rates in two-fluid hydrodynamics<sup>7</sup> or intranuclear cascade.8,12

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