One temperature step away from the critical point

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(Received 14 March 1996)

The disappearance of a wide range of critical fluctuations following a sudden temperature step away from the critical point is investigated theoretically. The step switches off the strong interaction on length scales larger than the final state correlation radius. This results in a nonequilibrium free field with fluctuations much larger than those in the final equilibrium state. In the course of relaxation, these initial non-Gaussian fluctuations decay sequentially on increasing scales, approaching the Gaussian equilibrium distribution. For an adiabatically insulated system, the theory predicts a power-law approach of the temperature to the new equilibrium. [S1063-651X(96)07609-X]

PACS number(s): 05.70.Jk, 05.70.Ln, 82.20.Mj

I. INTRODUCTION

Materials in near-critical conditions have unique fluctuational and thermodynamic features. The scaling theory and the method of renormalization group [1–4] explain the observed singularities of thermodynamic and kinetic characteristics and of the long-range correlations in equilibrium near-critical states. Recently, new experimental opportunities, especially those provided by the microgravity conditions, made it possible to investigate nonequilibrium near-critical states [5–7]. The slow thermal equilibration and large relaxation times of long-range correlations allow one to substantially perturb the local equilibrium by a rapid change of temperature T, pressure P, and/or other parameters. In a small but macroscopic element of the material, changes may be considered as homogeneous.

The critical point is manifested by large fluctuations of the order parameter (magnetic moment in magnets, particle density in liquid-gas systems, concentration in binary solutions, etc. [2,3]). The critical fluctuations are strongly correlated at length scales r ranging from atomic ($r_0 \sim 10^{-7}$ cm) up to the length scale of the correlation radius r_c. The spectrum of relaxation times $\tau(r)$ of these fluctuations ranges from microscopic times $\tau(r_0) \sim 10^{-12}-10^{-13}$ s up to the relaxation time $\tau(r_c)$. Both r_c and $\tau(r_c)$ increase as the system nears the critical temperature T_c. We consider a system with conserved [2,4] order parameter, e.g., the liquid-gas critical point at the critical isochore, or a binary solution critical point. In the scaling theory [1–4], the correlation radius yields the power law $r_c = r_0 \epsilon^{-\nu}$, and the relaxation time at the length scale r depends on the scale $\tau(r) = \tau(r_0)\epsilon^{-(3\nu+z)}$, where $\epsilon = (T-T_c)/T_c$ is the reduced temperature. The critical exponents $\nu > 0$ and $z \geqslant 0$ depend on the universality class of the system. Recently, the ZENO experiment [7] has set a benchmark $\epsilon \sim 10^{-7}-10^{-8}$, $r_c \sim 1-10$ μm, and $\tau(r_c) \sim 10^{1}-10^{3}$ s; this was achieved under microgravity conditions. One is able to perform a spatially homogeneous temperature change δT (resulting from, e.g., a rapid pressure shift) in a time small compared to $\tau(r_c)$. A relatively small sudden change of temperature may strongly perturb the local equilibrium on a wide range of scales, resulting in a near-critical transient state of the large scale fluctuations.

In the Ornstein-Zernicke approximation, rapid temperature changes in a near-critical system were first considered by Fixman [8]. The approximation neglects the strong interaction of critical ($r_0 \ll r < r_c$) fluctuations. This strong interaction of large scale fluctuations is described by the scaling theory and the renormalization group (RG) method [1–4].

The relaxation following a small perturbation ($|\delta\epsilon| \ll \epsilon$, $\delta\epsilon = \epsilon_{\rm fin} - \epsilon_{\rm in}$) has been studied using the RG method [2–4]. Here and below, the subscripts fin and in label the characteristics of the final and the initial ($t < 0$) states of the system. If the perturbation is strong, one has to study separately a step towards the critical point ($r_{c,\rm fin} \gg r_{c,\rm in}$) in which a new range of strongly interacting fluctuations appears, and a step away from the critical point ($r_{c,\rm fin} \ll r_{c,\rm in}$) where a large range of critical fluctuations disappears. In this paper, we study the latter case, a sudden large step away from the critical point.

II. DISCUSSION OF THE MODEL

The appearance, following a sudden temperature change, of a transient nonequilibrium state is a general phenomenon. Some details of the relaxation kinetics may depend on the dynamic universality class of the critical system. We assume that the time $\tau_{\rm ch}$ of the temperature change is short when compared to the relaxation times of long-range fluctuations, but $\tau_{\rm ch}$ is much larger than the relaxation time $\tau(r_0)$ at the atomic length scale r_0. There is then a range of small length scales for which the temperature change may be treated as quasistatic. The small scale degrees of freedom constitute the largest part of the degrees of freedom of the material, playing the role of a ''thermal bath'' for large scales. At times $t \gg \tau(r_0)$ one characterizes this ''thermal bath'' by a time-dependent temperature $T(t)$ and a reduced temperature $\epsilon(t) = [T(t) - T_c]/T_c$. The relaxation on large scales is slow, so that on the corresponding time scale one may treat the temperature change as a step at a given time $t_{\rm ch}$, chosen as $t_{\rm ch} = 0$. We consider a system described by a conserved real scalar order parameter. Systems belonging to this universality class are liquid-gas critical points and binary solution critical points.

To describe the fluctuations at different length scales, one represents the field $\phi(\mathbf{r},t)$ as a sum of Fourier harmonics:

$$\phi(r,t)=\sum_{k=0}^{k_\lambda}\phi_k e^{-ikr}. \tag{1}$$

The Fourier amplitudes ϕ_k yield the condition $\phi_k=\phi^*_{-k}$. A variable cutoff length $\lambda=2\pi/k_\lambda$ is introduced in (1) following the Wilson-Kadanoff RG method [1,2]. For a system at thermodynamic equilibrium, the probability $dW_{\mathrm{eq},\lambda}=w_{\mathrm{eq},\lambda}(\phi)D\phi(\mathbf{r})$ to find the system in an element $D\phi$ of the configuration space is defined by the effective Hamiltonian H_λ [1,2]:

$$w_{\mathrm{eq},\lambda}(\phi)D\phi=e^{[F_\lambda-H_\lambda(\phi)]/k_BT_c}D\phi,$$

$$F_\lambda=-k_BT_c\ \ln\int e^{-H_\lambda/k_BT_c}D\phi, \tag{2}$$

$$H_\lambda=-F_{\mathrm{reg}}-k_BT_c\ \ln\sum_{[\phi(r)]}e^{-H^{(\mathrm{mic})}/k_BT_c},\quad F_{\mathrm{reg}}=F-F_\lambda.$$

In the definition (2), $H^{(\mathrm{mic})}$ is the microscopic Hamiltonian of the system and k_B is the Boltzmann constant. The equilibrium free energy F depends on temperature and pressure but not on the configuration of the order parameter field; F_λ and F_{reg} are the singular and the regular parts of F, respectively [2]. The sum in (2) is over states of the system yielding the condition of given $\phi(\mathbf{r})$.

The order parameter field $\phi(\mathbf{r})$ defined by (1) represents only a small fraction of the degrees of freedom of the system, while most of these characteristics describe the details of the small length scales ($r<\lambda$). The definition of the effective Hamiltonian $H_\lambda(\phi,\epsilon)$ assumes an average over small scale degrees of freedom. Up to a regular function of ϵ, $H_\lambda(\phi,\epsilon)$ is the free energy of the system in the state in which all degrees of freedom have equilibrated except for the given large scale field $\phi(\mathbf{r})$.

We consider the simple standard model of a one-mode fluctuation kinetics of a conserved scalar order parameter field $\phi(\mathbf{r},t)$ [2,4], with the Langevin equations having the form

$$\frac{\partial\phi}{\partial t}=-\Delta\Gamma_\lambda\left(-\frac{\delta H_\lambda}{\delta\phi}+f_{\lambda,\mathrm{ext}}\right). \tag{3}$$

The external random force $f_{\lambda,\mathrm{ext}}(\mathbf{r},t)$ models the ''thermal bath.'' The kinetic operator Γ_λ (the kinetic coefficient in the simplest model) and the effective Hamiltonian H_λ depend on the cutoff length λ. The dynamic renormalization group (DRG) gives a method to find the changes in Eq. (3) when λ is increased from an old value λ_1 to a new value $\lambda_2>\lambda_1$. The necessary condition of applicability of DRG is thermal equilibrium at length scales $\lambda<\lambda_2$. At scales larger than λ_2, the equilibrium is not required, the relaxation on these scales is described by the Langevin equations of the theory, and the form of these equations is determined by the equilibrium on scales $R\leqslant\lambda$. The calculation below comes from this observation. One expects that at any given time $t>\tau_{\mathrm{ch}}$ there will be a length scale $\lambda_{\mathrm{eq}}(t)$ such that the fluctuations on scales $\lambda<\lambda_{\mathrm{eq}}(t)$ have approached equilibrium. The relaxation time for a conserved mode increases with length scale, so the equilibration length $\lambda_{\mathrm{eq}}(t)$ increases in time.

The main suggestion in the above scenario is that the relaxation in the nonequilibrium near-critical state is described by the Langevin equation (3) with the external random force $f(\mathbf{r},t)$ having the same properties as in the equilibrium state. This may be thought of as an approximation that may fail for large perturbations of equilibrium. An alternative suggestion is that the perturbation changes the external force ensemble. An experimental evidence of changes in the ensemble of external random force due to nonequilibrium was recently found in a noncritical liquid [9]. The test of the theory based on the above scenario will then prove the main suggestion.

The suggested scenario of relaxation allows one to employ the results of DRG calculations [1–4]. At a given time t, one chooses the cutoff length $\lambda<\lambda_{\mathrm{eq}}(t)$. The relaxation on scales $r>\lambda$ then yields the DRG equations determined by the effective Hamiltonian $H_{\lambda,\mathrm{fin}}$ and the kinetic operator $\Gamma_{\lambda,\mathrm{fin}}$ of the final equilibrium state; the initial conditions are defined by the initial state. This states a mathematical model of relaxation of large-scale fluctuations following a rapid temperature change, studied in the next section.

The relaxation of the nonequilibrium state releases the excess energy that is then converted to heat. To maintain the temperature $T(t)=\mathrm{const}$, one couples the internal ''heat bath'' provided by the small scale fluctuations to an external thermostat. Another experimental opportunity is to adiabatically insulate the system at $t>t_1>0$. The released energy then increases the temperature of the internal ''heat bath.'' At any time t, the rate of this heating is determined by the relaxation at the scales $r>\lambda_{\mathrm{eq}}(t)$, and therefore for the ''heat bath'' the slow variation of the temperature $T(t)$ may be treated as a quasistatic process.

III. THE DECAY OF CORRELATIONS FOLLOWING A STEP AWAY FROM THE CRITICAL POINT

For a temperature step away from the critical point, the final equilibrium state of the system is farther from the critical point than the initial one. The correlation radius $r_{c,\mathrm{fin}}$ in the final state is smaller than $r_{c,\mathrm{in}}$; a large step corresponds to $r_{c,\mathrm{fin}}\ll r_{c,\mathrm{in}}$. It is a known feature of the RG that in the critical range of scales $r<r_c$ the fluctuation ensemble has, up to small corrections, the same characteristics as in the critical point. The fluctuations are then expected to change significantly only on scales $r\geqslant r_{c,\mathrm{fin}}$. This range includes the scales $r_{c,\mathrm{fin}}<r<r_{c,\mathrm{in}}$ belonging to the critical range of scales of the initial state.

According to the above suggestion, at $t>\tau(r_{c,\mathrm{fin}})$ the fluctuations on scales $r\leqslant\lambda(t)$, $\lambda(t)\gg r_{c,\mathrm{fin}}$ approach the new equilibrium state. The application of the DRG then gives the effective Hamiltonian and the Langevin equations for the fluctuations on the length scales $r>\lambda$. As described in the DRG [1–4], on length scales $\lambda\gg r_{c,\mathrm{fin}}$ at times $t\gg\tau(r_{c,\mathrm{fin}})$ the system approaches the Gaussian fixed point. The Langevin equations then have the form (3) with the kinetic coefficient $\Gamma=\lim_{\lambda\to\infty}\Gamma_\lambda$ depending on ϵ_{fin} but not on the scale λ, and the effective Hamiltonian having the form of a free field Hamiltonian [1–3]

$$H_\lambda=\sum_{k<k_\lambda}H_k,\quad H_k=\frac{1}{2\chi_{\mathrm{fin}}}|\phi_k|^2. \tag{4}$$

Here, $\chi_{\rm fin}\equiv\lim_{\lambda\to\infty}\chi_{\lambda,\rm fin}\sim\epsilon_{\rm fin}^{-\nu(2-\eta)}$ is the macroscopic susceptibility of the system in the final state (the isothermal compressibility for the liquid-gas system). In the range of scales considered ($r>r_{c,\rm fin}$) $\chi_{\rm fin}$ does not depend on the wave vector $\mathbf{k}$.

The reduction of the problem to that of a free field allows one to solve the problem of long-range fluctuations without further assumptions. The fluctuations at length scales larger than r_c are strongly interacting only with the critical range ($r<r_c$) fluctuations but not with each other. Each Fourier harmonics $\phi_k(t)$ yields now a separate equation one derives from (3) and (4):

$$\frac{d\phi_k}{dt}=-\Gamma k^2\left[\frac{\phi_k}{\chi_{\rm fin}}+f_{k,st}(t)\right]. \tag{5}$$

The force $f_{k,st}(t)$ in (5) is δ correlated in time, and normalized according to the fluctuation-dissipation theorem [2,4]. The quantity $D_{\rm fin}=\Gamma/\chi_{\rm fin}$ is the final state diffusion coefficient, and Eq. (5) describes a linear diffusion process in the presence of a fluctuating current $j=-\Gamma\ \mathrm{grad}\, f_{\rm st}$. The general solution to the linear equations (5) may be written as

$$\phi_k(t)=\phi_k(0)e^{-t/\tau_k}+\phi_{k,st}(t),\quad \tau_k=\frac{1}{D_{\rm fin}k^2}, \tag{6}$$

where the first term is the solution $\phi_{k,h}(t)$ of the corresponding homogeneous equations, and $\phi_{k,st}$ describes the motion induced by the external force $f_{st}(t)$. The initial conditions are determined by the initial equilibrium state in which the level of fluctuations was much higher than in the final state. At $t<\tau_k$, $\langle|\phi_k|^2\rangle\approx\langle|\phi_{k,h}|^2\rangle\gg\langle|\phi_{k,st}|^2\rangle$.

The statistical properties of the fluctuations may be described with the help of irreducible correlation functions [2,10] $G_n(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\ldots,\mathbf{k}_n;t)=\langle\langle\phi(\mathbf{k},t)\phi(\mathbf{k}_1,t)\phi(\mathbf{k}_2,t)\cdots\phi(\mathbf{k}_n,t)\rangle\rangle_t$ defined as

$$\begin{aligned}G_n(k_1,k_2,\ldots,k_n;t)&\equiv\langle\langle\phi_{k_1}(t)\phi_{k_2}(t)\cdots\phi_{k_n}(t)\rangle\rangle\\&\equiv\frac{\delta^n}{\delta\omega(k_1)\delta\omega(k_2)\cdots\delta\omega(k_n)}\ln Z|_{\omega=0},\\ Z\equiv\langle e^{\int\omega(k)\phi_k(t)dV}\rangle&\equiv\int e^{\int\omega(k)\phi_k(t)dV}DW[\phi_k;t].\end{aligned} \tag{7}$$

Here, $Z[\omega(\mathbf{k})]$ is the generating functional [2,10]. The averages $\langle\ \rangle$ and irreducible averages $\langle\langle\ \rangle\rangle$ in (7) are over the instantaneous statistical ensemble. In the first equation in (7), one sets $\omega(\mathbf{k})=0$ after the functional derivatives are taken. Due to the space homogeneity of the system, the functions $G_n(\mathbf{k}_1,\ldots,\mathbf{k}_n;t)$ may differ from zero only if $\mathbf{k}_1+\mathbf{k}_2+\cdots+\mathbf{k}_n=0$. At $t=0$, the irreducible correlation functions G_n yield the scaling laws given by the scaling theory [2,11]; for all arguments k in the critical range of scales of the initial state $k_1\sim k_2\sim\cdots\sim k_n\sim k$, $kr_{c,\rm in}\gg1$, one finds $G_n\sim k^{n(-2+\eta)/2}$. The irreducible correlations G_n for $n>2$ characterize the deviation of the ensemble from the Gaussian form. In the initial state, the interaction is strong and the fluctuations are non-Gaussian on scales $r<r_{c,\rm in}$. As discussed above, the temperature step switches off the strong interaction in the range of scales $r_{c,\rm in}>R>r_{c,\rm fin}$; in the final state $G_n=0$ for $n>2$. The Gaussian part $\phi_{k,st}$ does not contribute to G_n with $n>2$. One obtains then for $n>2$

$$\begin{aligned}G_n(k_1,\ldots,k_n;t)&=G_n(k_1,\ldots,k_n;0)e^{-D_{\rm fin}(k_1^2+k_2^2+\cdots+k_n^2)t},\\ G_n(k_1,k_2,\ldots,k_n;0)&\equiv\langle\langle\phi_{k_1}\cdots\phi_{k_n}\rangle\rangle_{\rm in}.\end{aligned} \tag{8}$$

The function $G_2(k,-k)=\langle\langle|\phi_k(t)|^2\rangle\rangle$ describes the width of the probability distribution of the harmonics $\phi_k(t)$:

$$\begin{aligned}\langle\langle|\phi_k|^2\rangle\rangle_t&=\langle\langle|\phi_k(t)|^2\rangle\rangle=(\langle\langle|\phi_k|^2\rangle\rangle_{\rm in}-\langle\langle|\phi_k|^2\rangle\rangle_{\rm fin})e^{-t/\tau_k}\\&\quad+\langle\langle|\phi_k|\rangle\rangle_{\rm fin},\\ \langle\langle|\phi_k|^2\rangle\rangle_{\rm in}&\sim k^{-2+\eta},\quad r_{c,\rm in}\gg\frac{1}{k}\gg r_{c,\rm fin}.\end{aligned} \tag{9}$$

The correlation functions $G_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;t)$ in coordinate space are related to $G_n(k_1,\ldots,k_n;t)$ by the formula

$$\begin{aligned}&G_n(r_1,r_2,\ldots,r_n;t)\\&\quad=\sum_{k_1+k_2+\cdots+k_n=0}\langle\langle\phi_{k_1}(t)\phi_{k_2}(t)\cdots\phi_{k_n}(t)\rangle\rangle\\&\qquad\times e^{-i(k_1r_1+k_2r_2+\cdots+k_nr_n)}.\end{aligned} \tag{10}$$

On a given length scale $r=1/k$, the ensemble of fluctuations is then non-Gaussian at times $t\ll t_G\sim\tau(k)\ln A_k$, when the fluctuations are still large compared to the final equilibrium. At $t>t_G$, the irreducible correlation functions G_n for $n>2$ become small while G_2 approaches the final equilibrium value. The ensemble then approaches the Gaussian form.

IV. THE ENERGY OF THE RELAXATION

A rapid increase of the temperature requires more energy than a slow heating. The effective Hamiltonian $H_\lambda(\phi)$ is by definition [see formula (2)] the free energy in a state with given ϕ_k, $k<1/\lambda$. The entropy in this state is then $S_\lambda=-k_b[\partial H_\lambda(\phi,\epsilon)/\partial T]=-k_b(1/T_c)\partial H_\lambda/\partial\epsilon$. From (3) one finds the contribution $s_k=k_b(1/T_c)\langle H_k\rangle(\partial\ln\chi/\partial\epsilon)$ of a single harmonic ϕ_k to the entropy. The single harmonic internal energy U_k is (according to the thermodynamic formula $U=F+TS$) $U_k=\langle H_k\rangle+T_cs_k\approx T_cs_k$. Let the initial state be $\epsilon_{\rm in}=0$, $r_{c,\rm in}=\infty$, $\langle|\phi_k|^2\rangle_{\rm in}\sim k^{-2+\eta}$ [1–3]. In the final equilibrium state one finds $\langle|\phi_k|^2\rangle_{\rm fin}\sim\chi(\epsilon_{\rm fin})$, $k\ll1/r_{c,\rm fin}$. Summing up the contributions from all large-scale harmonics, one obtains the excess energy $U_{\rm stor}$ stored at $t\sim\tau(r_{c,\rm fin})$:

$$\begin{aligned}U_{\rm stor}(0)&=\frac{V}{2}\frac{\partial\ln\chi_{\rm fin}}{\partial\epsilon}\frac{1}{\chi_{\rm fin}}\int_0^{1/\lambda}\langle|\phi_k|\rangle_{\rm in}^2\frac{d^3k}{(2\pi)^3}\\&\approx Nk_bT_c\epsilon_{\rm fin}^{3\nu-1}.\end{aligned} \tag{11}$$

Here $\lambda\sim r_{c,\rm fin}$, and $N\sim V/r_0^3$ is the number of molecules in the system. At $t>\tau(r_\lambda)$ the stored energy is gradually converted into heat. The released heat $Q(t)=U_{\rm stor}(0)-U_{\rm stor}(t)$ as a function of time is

$$Q(t)=\frac{V}{2}\frac{\partial \ln \chi}{\partial \epsilon_{\rm fin}}\frac{1}{\chi_{\rm fin}}\int_0^{k_\lambda}[\langle|\phi_k(0)|^2\rangle-\langle|\phi_k(t)|^2\rangle]\frac{d^3k}{(2\pi)^3}$$

$$\propto U_{\rm stor}(0)\left[1-\left(\frac{\tau(r_{c,\rm fin})}{t}\right)^{(1+\eta)/2}\right]. \quad (12)$$

So far we have considered the relaxation in a system where the temperature $T(t)$ is maintained constant during the relaxation, so that at any time $t>0$, $\epsilon(t)=\epsilon_{\rm fin}$. If, instead, one adiabatically insulates the system at times $t>\tau(r_{\rm fin})$, the time-dependent temperature $T(t)$ yields the relation $dQ=C_v dT=C_v T_c d\epsilon(t)$, with $C_v\sim\epsilon_{\rm fin}^{3\nu-2}$ being the heat capacity at constant volume. The approach of the temperature to equilibrium follows now from the form (12) of the heat release. For $t>\tau(r_{c,\rm fin})$ one obtains

$$\epsilon(t)-\epsilon_{\rm fin}\sim\epsilon_{\rm fin}[\tau(r_{c,\rm fin})/t]^{\zeta_+},\quad \zeta_+=(1+\eta)/2. \quad (13)$$

The RG prediction, confirmed by experiments [11], for η is $\eta\approx0.033$. The new critical exponent $\zeta_+\approx0.517$ is numerically close to the value 1/2 one obtains in the mean field (Landau) theory.

V. CONCLUSION DISCUSSION

A rapid temperature step away from the critical point triggers a hierarchical relaxation of long-range fluctuations. The short time equilibration of small scale ($r<r_{c,\rm fin}$) fluctuations leads to an increase of the nonequilibrium free energy of large scale ($r>r_{c,\rm fin}$) fluctuations due to a decrease of the susceptibility, while the large scale configuration remains unchanged. The interaction of fluctuations inside the range $r>r_{c,\rm fin}$ becomes weak; the interaction of large scale fluctuations with those in the range $r<r_{c,\rm fin}$ remains strong and results in the singular dependence of the susceptibility $\chi_{\rm fin}$ and of the mobility $\Gamma_{\rm fin}$ on $\epsilon_{\rm fin}$. In the final state, a wide range of initial critical fluctuations disappears. A step toward the critical point would lead to the appearance of such a range [12].

In the RG, the strong interaction of fluctuations on a given length scale is substantially renormalized by their interaction with smaller length scales. One considers [1,2] a Hamiltonian $H_\lambda=H^*+\delta H_\lambda$ that has a small deviation δH_λ from the fixed point. At larger scales λ, this deviation results in a crossover from a non-Gaussian to a Gaussian fixed point. In this paper, the perturbation δH_λ of the Hamiltonian is a dynamic one, and the equilibration on large length scales is a hierarchical relaxation process.

The scenario considered in this paper assumes the ensemble of external random forces unchanged by the perturbation. The predicted decay of correlations may be tested in scattering experiments. Recently, another promising way to study the fluctuation ensemble was proposed, based on a direct visualization of the fluctuation picture [13].

Temperature ''tails'' are predicted for the temperature relaxation in an adiabatically insulated system; the temperature approaches its final equilibrium value from below by a power law.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge R. W. Gammon, V. L. Pokrovsky, M. A. Ratner, and R. Allen Wilkinson for helpful discussions. This study was supported by NASA Grant No. NAG3-1617.

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