

Energy-loss spectrum for inelastic scattering of charged particles in disordered systems near the critical point

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The behavior of the theoretically predicted correlational "fine" energy-loss spectrum of inelastic electron scattering in disordered systems close to single resonance is investigated near the critical point. In extending our earlier work, it is shown that the relation of the statistical expression of the cross section of energy loss to the function which describes the line shape in an ideal gas asymptotically increases near the critical point as a power law. "Fracton" interpretation of display of the localization of a single excitation in disordered systems in the resonance-line shape of the energy-loss spectrum is suggested. The possibility of direct determination of the pair distribution function (without Fourier transformation of the structure factor) using the method of charged-particle scattering is discussed.

I. INTRODUCTION

The development of the technology of inelastic electron scattering experiments¹ gives some hope to the study of the behavior of the energy-loss spectrum in disordered systems close to single resonance (with width $\sim 10^{-1}$ eV), out of the Doppler width. In this region, it was theoretically predicted² that a statistical "fine" structure of the energy-loss spectrum due to many-particle intermolecular correlation would appear.

In this paper we will extend earlier work² about the shape of the resonance in the energy-loss spectrum on the critical point, where the role of many-particle correlations is particularly large. Moreover, in connection with the appearance of ideas of geometric interpretation of the scaling phenomena with the help of the introduction of self-similar fractal objects, the study of the correlational structure of the atomic line shapes, which are due to interatomic correlations, by the correlational expansion method (which is based on cluster expansions), is a good method for distinguishing the dynamics of the fractal objects, particularly near the critical point. This question will be discussed in Secs. V and VI of this paper.

II. CONSTRUCTION OF TASK (CORRELATIONAL EXPANSION FOR CROSS SECTION OF ENERGY LOSS)

Consider the scattering of a monochromatic beam of charged particles (electrons with energy $E \sim 10^3$ eV) by a disordered system of N identical, symmetrical molecules with volume V and temperature $T \sim 10^2$ K. Let the average velocity of the incident electrons be at least one order larger than the velocities of the bound charges of the molecules. For this case, the cross section of energy-loss scattering may be calculated by Born's approximation. Finally, the target molecules can be considered as being at rest during the elementary act of scattering. The bound charges of the target molecules distribute not only in the region of localization of one single molecule, but also together with the other molecules over the entire volume occupied by the system. Therefore, the expression for the cross section of energy loss must be averaged over all the molecular configurations with the Gibbs distribution function. The resulting quantum-mechanical expression for the doubly differential cross section of scattering $d^2\tau/d\theta d\epsilon$ (or the cross section of energy losses) is

$$I_\theta(\epsilon) \equiv \frac{d^2\tau}{d\theta d\epsilon} = \frac{1}{V} \left[\frac{e}{E\theta^2} \right]^2 \left\langle \sum_{(\alpha)} |\rho_{10,\alpha}^{(N)}(\mathbf{q}; \{\mathbf{R}\})|^2 \delta(E_{1\alpha}(\{\mathbf{R}\}) - E_0(\{\mathbf{R}\}) - \epsilon) \right\rangle, \quad (1)$$

where $\rho_{10,\alpha}^{(N)}(\mathbf{q}, \{\mathbf{R}\})$ is the Fourier transformation of the matrix element from the charge-density operator, $\hbar\mathbf{q}$ is the impulse of scattering (i.e., the change of the electron impulse by scattering), $\{\mathbf{R}\} \equiv \mathbf{R}_1, \dots, \mathbf{R}_N$ are the coordinates of the center of mass of the N molecules, $E_{1\alpha}(\{\mathbf{R}\})$

and $E_0(\{\mathbf{R}\})$ are, respectively, the energy of the final- and the initial-system states in the adiabatic approximation, the sum from α goes over all the excited states of the system which gives rise to distinctive levels of isolated molecules, ϵ is the electron energy loss due to scattering,

$\delta(\dots)$ is the Dirac δ function, the angular brackets $\langle \dots \rangle$ mean the procedure of averaging over all the molecular configurations with the Gibbs distribution function, and θ is the scattering angle. Expression (1) is the generalization of the famous expression for the cross section of charged-particle inelastic scattering,³ in the case where we consider our condensed system as a single large molecule.

Consider the small-angle scattering with an energy loss near the energy $\hbar\omega_1$, which corresponds to the isolated excitation of a single molecule of the system. We shall investigate the singularities in the distribution of electrons inelastically scattered by energy losses $I_\theta(\epsilon)$, in the nar-

row ($\epsilon \sim 10^{-2} - 10^{-1}$ eV) neighborhood of distinctive resonance $\hbar\omega_1$ on a fixed scattering angle $\theta \leq 1^\circ$.

We want to emphasize that the widening of the line in the energy-loss spectrum due to molecular motion is approximately $\Delta \sim (mk_B TE\theta/M)^{1/2}$,⁴ where m and M are the masses of electron and molecule, respectively, and k_B is the Boltzmann constant. Thus, for the parameter values of our problem, $\Delta \sim 10^{-2} - 10^{-3}$ eV.

Following Ref. 2, let us expand $I_\theta(\epsilon)$ in a correlation series, every part of which describes the contribution to the total cross section of energy loss due to scattering by fluctuational complexes of isolated s particles ($s = 1, N$),

$$I_\theta(\epsilon) = a'(\theta) \sum_s \frac{n^s}{s!} \int \cdots \int \left[\sum_{\alpha=1}^{\nu_s} Z_{\alpha,s}(\mathbf{q}; \mathbf{R}_1, \dots, \mathbf{R}_s) \delta(\epsilon + \mu_{\alpha,s}(\mathbf{R}_1, \dots, \mathbf{R}_s)) \right. \\ - \frac{s!}{(s-1)!} \sum_{\alpha=1}^{\nu_{s-1}} Z_{\alpha,s-1}(\mathbf{q}; \mathbf{R}_1, \dots, \mathbf{R}_{s-1}) \delta(\epsilon + \mu_{\alpha,s-1}(\mathbf{R}_1, \dots, \mathbf{R}_{s-1})) \\ + \frac{s!}{(s-2)!2!} \sum_{\alpha=1}^{\nu_{s-2}} Z_{\alpha,s-2}(\mathbf{q}; \mathbf{R}_1, \dots, \mathbf{R}_{s-2}) \delta(\epsilon + \mu_{\alpha,s-2}(\mathbf{R}_1, \dots, \mathbf{R}_{s-2})) \\ \left. - \cdots - (-1)^s Z_1(\mathbf{q}; \mathbf{R}_s) \right] g_s(\mathbf{R}_1, \dots, \mathbf{R}_s) d\mathbf{R}_1 \cdots d\mathbf{R}_s, \quad (2)$$

where the sum from α equals 1 to ν_s goes over all the states of s -particle complexes; $n = N/V$ the number density particles;

$$a'(\theta) = \left(\frac{e}{E\theta^2} \right)^2,$$

$$Z_{\alpha,s}(\mathbf{q}; \mathbf{R}_1, \dots, \mathbf{R}_s) \equiv |\rho_{10,\alpha}^{(s)}(\mathbf{q}; \mathbf{R}_1, \dots, \mathbf{R}_s)|^2;$$

$\rho_{10,\alpha}^{(s)}(\mathbf{q}; \mathbf{R}_1, \dots, \mathbf{R}_s)$ is the Fourier transformation of the matrix element from the charge-density operator; $g_1(\mathbf{R}_1), g_2(\mathbf{R}_1, \mathbf{R}_2), g_3(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3), \dots, g_s(\mathbf{R}_1, \dots, \mathbf{R}_s)$ accordingly are the one-, two-, three-, etc., s -particle distribution functions;⁵ and $\mu_{\alpha,s}(\mathbf{R}_1, \dots, \mathbf{R}_s)$ are the shifts of the energy of single excitation due to intermolecular interactions in s -particle complexes. The expression in large parentheses in (2) describes irreducible s -particle contributions in the cross section of energy loss for inelastic electron scattering with fixed parameters in disordered systems. Every contribution in expression (2) is a functional of density and temperature because all the functions $\{g_s\}$ are functionals of these parameters.

III. STATISTICAL MODEL OF LOCALIZED EXCITATION IN DISORDERED SYSTEMS

Consider the model in which the excitation is localized

on the single molecule and the probability of exchanges of the excitation between molecules in complexes is negligible. This situation is adequate for considering dipole-forbidden excitation, when resonance interaction between an excited molecule and a molecule in the ground state decreases with interatomic distance R more rapidly than R^{-3} . We shall suppose that the excited level of a single molecule is degenerated. Then, in the zeroth order of the perturbation theory on interatomic interaction, where exchange effects are negligible, we obtain

$$Z_{\alpha,s}(\mathbf{q}, \mathbf{R}_1, \dots, \mathbf{R}_s) \simeq Z_1(\mathbf{q}) \equiv |\rho_{10}^{(1)}(\mathbf{q})|^2, \quad \alpha = s \quad (3)$$

$$\mu_{\alpha,s}(\mathbf{R}_1, \dots, \mathbf{R}_s) = \sum_{j=1}^{s-1} \mu(|\mathbf{R}_j - \mathbf{R}_\alpha|),$$

where we put $\mathbf{R}_s = 0$; $\mu(R)$ is the exchange of energy of the single excitation in a pair of molecules which shifts from one to the other on vector \mathbf{R} ; α , in this case, means that the excitation is localized at the molecule with number α . Due to the symmetry of the functions $\{g_s\}$, with help from the Fourier reducing of the Dirac δ function, we can rewrite expression (2) as

$$I_\theta(\epsilon) = a(\theta) Z_1(\mathbf{q}) \sum_{s=1}^{\infty} \frac{n^s}{(s-1)!} \int_{-\infty}^{\infty} d\tau \exp(-i\epsilon\tau) \int \cdots \int \prod_{j=1}^{s-1} \{\exp[-i\mu(\mathbf{R}_j)\tau] - 1\} g_s(\mathbf{R}_1, \dots, \mathbf{R}_{s-1}) d\mathbf{R}_1 \cdots d\mathbf{R}_{s-1}, \quad (4)$$

$$g_s(\mathbf{R}_1, \dots, \mathbf{R}_{s-1}) \equiv g_s(\mathbf{R}_1, \dots, \mathbf{R}_{s-1}, 0), \quad a(\theta) \equiv \frac{a'(\theta)}{2\pi}.$$

Thus, if $\mu(R)$ is a finite function [i.e., $\mu(R)$ equals zero outside a sphere with radius r_0], integration in (4) goes over the volume of the sphere $V_0 = (4\pi/3)r_0^3$ with its centrum in the origin of the coordinate system. The series (4) for the finite function $\mu(R)$ is actually generated in the finite sum (in the sphere with radius $r_0 < \infty$, we can only put a finite number of molecules); this means that, starting with some number s_0 , all the functions $\{g_s\}$ ($s > s_0$), turn into zero for all $R_s \geq r_0$. In order to get rid of the proof of convergence of the correlation series in the case that $\mu(R)$ is not finite, but only a rapidly decreasing function, it is sufficient to introduce under the sign of the in-

tegral in (4) the cutoff multiplier $\exp(-\Delta^2\tau^2)$ (it is necessary for including the natural broadening of spectral lines due to molecular motion and the effect of back-scattering). A more complete consideration of this problem was given in Ref. 2.

IV. CORRELATIONAL STRUCTURE OF RESONANCE-LINE SHAPE (CSR)

For distinguishing the correlational contributions in the cross section of energy loss, we shall write the s -particle distribution function in the form of a series of irreducible contributions,

$$g_s(\mathbf{R}_1, \dots, \mathbf{R}_{s-1}) = 1 + \sum_{i=1}^s \sum_{j=i+1}^s h_2(\mathbf{R}_i - \mathbf{R}_j) + \sum_{i=1}^s \sum_{j=i+1}^s \sum_{k=j+1}^s h_3(\mathbf{R}_i - \mathbf{R}_j; \mathbf{R}_i - \mathbf{R}_k) + \dots + h_s(\mathbf{R}_1, \dots, \mathbf{R}_{s-1}), \quad (5)$$

where $\{h_s(\mathbf{R}_1, \dots, \mathbf{R}_{s-1})\}$ is the set of irreducible correlation functions,

$$\begin{aligned} h_2(\mathbf{R}) &= g_2(\mathbf{R}) - 1, \\ h_3(\mathbf{R}_i, \mathbf{R}') &= g_3(\mathbf{R}, \mathbf{R}') - g_2(\mathbf{R}) - g_2(\mathbf{R}') - g_2(\mathbf{R} - \mathbf{R}') + 2, \\ &\vdots \\ h_s(\mathbf{R}_1, \mathbf{R}_2, \dots, \mathbf{R}_{s-1}) &= g_s(\mathbf{R}_1, \dots, \mathbf{R}_{s-1}) - g_{s-1}(\mathbf{R}_1, \dots, \mathbf{R}_{s-2}) - \dots - (-1)^s(s-1). \end{aligned} \quad (6)$$

Let us rewrite expression (4), with the help of (5), and distinguish noncorrelation contributions and the corrections to them due to two-, three-, ..., s -particle correlations

We obtain the noncorrelational term $I_\theta^{(0)}(\epsilon)$, which describes the shift and the broadening of the spectral lines as a function of pressure in an ideal-gas approximation, if we put all $\{g_s\} = 1$ in (4), namely,

$$I_\theta^{(0)}(\epsilon) = a(\theta)Z_1(\mathbf{q})n \int_{-\infty}^{\infty} d\tau \exp \left[-i\epsilon\tau + 4\pi n \int_0^{\infty} (e^{-i\mu(R)\tau} - 1) R^2 dR \right] \equiv a(\theta)Z_1(\mathbf{q})n M_\theta^{(0)}(\epsilon). \quad (7)$$

If, for example we used in $\mu(R)$ only the long-range asymptote of this function, i.e., $\mu(R) = -C/R^6$ (if we consider the dipole-forbidden excitation), we immediately obtain the expression derived by Margenau and Watson⁶ and others,⁷ which describes the behavior of the spectral line of a molecule dissolved in an ideal gas,

$$I_\theta^{(0,M)}(\epsilon) = a(\theta)Z_1(\mathbf{q})n \lambda \epsilon^{-3/2} \exp \left[-\frac{\pi \lambda^2}{\epsilon} \right] = a(\theta)Z_1(\mathbf{q})n M_\theta^{(0,M)}(\epsilon), \quad \lambda \equiv \frac{2\pi}{3}(c)^{1/2}n. \quad (8)$$

In general, if we confine ourself only to the first two terms in (5), we obtain, with the help of (4),

$$\begin{aligned} I_\theta(\epsilon) &= a(\theta)Z_1(\mathbf{q})n \left[M_\theta^{(0)}(\epsilon) + n \int \Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R})) h_2(\mathbf{R}) d\mathbf{R} \right. \\ &\quad \left. + \frac{1}{2} n^2 \int \int [\Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R}) + \mu(\mathbf{R}')) - \Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R})) - \Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R}'))] \right. \\ &\quad \times h_2(\mathbf{R} - \mathbf{R}') d\mathbf{R} d\mathbf{R}' \left. \right] \equiv a(\theta)Z_1(\mathbf{q})n M_\theta^{(1)}(\epsilon), \\ \Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R})) &\equiv M_\theta^{(0)}(\epsilon + \mu(\mathbf{R})) - M_\theta^{(0)}(\epsilon). \end{aligned} \quad (9)$$

In the next approximation,

$$\begin{aligned} I_\theta(\epsilon) &= a(\theta)Z_1(\mathbf{q})n \left[M_\theta^{(1)}(\epsilon) + \frac{1}{2} n^2 \int \int \int [\Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R}) + \mu(\mathbf{R}')) - \Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R})) \right. \\ &\quad \left. - \Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R}'))] h_3(\mathbf{R}, \mathbf{R}') d\mathbf{R} d\mathbf{R}' \right], \end{aligned} \quad (10)$$

etc. In (9) and (10), $M_\theta^{(0)}(\epsilon)$ is, in general, determined by (7). Thus expressions such as (9) and (10) give the possibility for analytical (or numerical) calculation of corrections to the expressions (7) and (8) in any order of approximations on correlations.

Following Parceival's theorem, expressions (9), (10), and similar ones may be written in the form of a relation between the doubly differential cross section of inelastic electron scattering $I_\theta(\epsilon)$ and the static structure factors of the medium $\{S_j\}$. For example, using (9) and (10), we obtain

$$I_\theta(\epsilon) = a(\nu) Z_1(\mathbf{q}) n \left[M_\theta^{(0)}(\epsilon) + \int K_2(\epsilon, \mathbf{k}) h_2(\mathbf{k}) d\mathbf{k} + \int \int K_3(\epsilon, \mathbf{k}, \mathbf{k}') h_3(\mathbf{k}, \mathbf{k}') d\mathbf{k} d\mathbf{k}' \right], \quad (11)$$

where

$$\begin{aligned} K_2(\epsilon, \mathbf{k}) &= \frac{1}{(2\pi)^3} \int e^{i\mathbf{k}\cdot\mathbf{R}} \left[\Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R})) + \frac{1}{2} n \int e^{-i\mathbf{k}\cdot\mathbf{R}'} [\Delta M_\theta(\epsilon + \mu(\mathbf{R}) + \mu(\mathbf{R}')) - \Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R})) \right. \\ &\quad \left. - \Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R}'))] \right] d\mathbf{R} d\mathbf{R}', \end{aligned} \quad (12)$$

$$K_3(\epsilon, \mathbf{k}, \mathbf{k}') = \frac{n^2}{2(2\pi)^3} \int \int e^{i\mathbf{k}\cdot\mathbf{R} + i\mathbf{k}'\cdot\mathbf{R}'} [\Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R}) + \mu(\mathbf{R}')) - \Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R})) - \Delta M_\theta^{(0)}(\epsilon + \mu(\mathbf{R}'))] d\mathbf{R} d\mathbf{R}',$$

$$S_2(\mathbf{k}) = 1 + nh_2(\mathbf{k}), \quad S_3(\mathbf{k}, \mathbf{k}') = 1 + n[h_2(\mathbf{k}) + h_2(\mathbf{k}') + h_2(\mathbf{k} - \mathbf{k}')] + n^2 h_3(\mathbf{k}, \mathbf{k}'), \quad (13)$$

where $h_2(\mathbf{k})$, $h_3(\mathbf{k}, \mathbf{k}')$, etc. are the Fourier transforms of $\{h_s\}$ [see (6)]; $S_2(\mathbf{k})$, $S_3(\mathbf{k}, \mathbf{k}')$, etc., the static structure factors of medium; etc. Expressions of type (10) are important, because the static structure factors (13) may be measured in alternative experiments, for example, by elastic x-ray or neutron scattering.⁸ In other words, with the help of (12), we obtain a connection between the cross section of inelastic scattering and the static structure factors which may be measured in elastic x-ray or neutron scattering experiments.⁹

V. BEHAVIOR OF CSR NEAR THE CRITICAL POINT

The correlational series (4) allows us to make a sum up to any approximation of the many-particle distribution

functions [see (5)]. Let us investigate, for example, the behavior of single resonances in the energy-loss spectrum near the critical point, using for this aim the simplest Ornstein-Zernike correlational function $g_2(R)$,

$$g_2(R) = A \frac{e^{-xR}}{R}, \quad (14)$$

where A is a temperature-dependent coefficient with the dimension of a distance and x is the inverse correlational radius. From Eqs. (9) and (14), follows that, including at sufficiently moderate densities only the terms proportional to n , the function $I_\theta(\epsilon)$ is given by

$$\begin{aligned} I_\theta(\epsilon) &= a(\theta) Z_1(\mathbf{q}) n \left\{ M_0^{(0,M)}(\epsilon) + 4\pi An \lambda \left[\int_{(c/\epsilon)^{1/6}}^{\infty} (\epsilon - c/R)^{-3/2} \exp \left[-\frac{\pi\lambda^2}{\epsilon - \frac{c}{R^6}} - xR \right] R dR \right. \right. \\ &\quad \left. \left. - \epsilon^{-3/2} \exp \left[-\frac{\pi\lambda^2}{\epsilon} \right] \int_{(c/\epsilon)^{1/6}}^{\infty} \exp(-xR) R dR \right] \right\}. \end{aligned} \quad (15)$$

In Ref. 2 it was shown that, being negligible in this case, the contribution in (9) due to rapid convergence of correlational series has another order of density and may be separated. After changing the variable in the first integral and after the calculation of the second integral, we can rewrite expression (15) as follows:

$$\begin{aligned} I_\theta(\epsilon) &= I_\theta^{(0,M)}(\epsilon) \left\{ 1 + 4\pi An \left[\frac{1}{6} c^{1/3} \epsilon^{1/6} \int_0^{\infty} y^{-4/3} \left(y + \frac{1}{\epsilon} \right)^{5/6} \exp \left[-\pi\lambda^2 y - \frac{xc^{1/6}}{\epsilon^{1/3}} \left(\frac{1}{y} + \epsilon \right)^{1/6} \right] dy \right. \right. \\ &\quad \left. \left. - x^2 \Gamma(2, x(c/\epsilon)^{1/6}) \right] \right\}, \end{aligned} \quad (16)$$

where $\Gamma(2, x(c/\epsilon)^{1/6})$ is the incomplete Γ function,¹⁰ in particular,

$$\Gamma(2, x(c/\epsilon)^{1/6}) = [1 + x(c/\epsilon)^{1/6}] \exp[-x(c/\epsilon)^{1/6}]. \quad (17)$$

Let us appraise now the behavior of $I_\theta(\epsilon)$ with the help of (16), for limiting values of ϵ . We define the integral $\Xi(\epsilon, n, x)$,

$$\Xi(\epsilon, n, x) \equiv \int_0^\infty y^{-4/3} \left[y + \frac{1}{\epsilon} \right]^{5/6} \exp \left[-\pi\lambda^2 y - \frac{xc^{1/6}}{\epsilon^{1/3}} \left(\frac{1}{y} + \epsilon \right)^{1/6} \right] dy .$$

When $\epsilon \rightarrow 0$, we can approximately calculate the value of Ξ by means of the method of steepest descent.¹¹ We obtain as a result

$$[\Xi(\epsilon, n, x)]_{\epsilon \rightarrow 0+} \approx \frac{63^{3/14}(2\pi)^{5/7}}{7^{1/2}} \lambda^{3/7} (xc^{1/6})^{-5/7} \epsilon^{-25/42} \exp \left[-\frac{7}{6} (6\pi c)^{1/7} \left(\frac{\lambda x^3}{\epsilon} \right)^{2/7} \right]. \quad (18)$$

Combination of (17), (18), and (16) yields

$$[I_\theta(\epsilon)]_{\epsilon \rightarrow 0+} \approx I_\theta^{(0,M)}(\epsilon) \left[1 + 4\pi An x^{-5/7} \left\{ \frac{(2\pi)^{4/14}}{7^{1/2} 3^{3/14}} c^{3/14} (\lambda\epsilon)^{-3/7} \exp \left[-\frac{7}{6} (6\pi c)^{1/7} \left(\frac{\lambda x^3}{\epsilon} \right)^{2/7} \right] \right. \right. \\ \left. \left. - x^{-2/7} \left(\frac{c}{\epsilon} \right)^{1/6} \exp \left[-x \left(\frac{c}{\epsilon} \right)^{1/6} \right] \right\} \right]_{\epsilon \rightarrow 0} \rightarrow I_\theta^{(0,M)}(\epsilon), \left[\frac{I_\theta(\epsilon)}{I_\theta^{(0,M)}(\epsilon)} \right]_{\epsilon \rightarrow 0} \rightarrow 1 . \quad (19)$$

In the opposite formal limit, when $\epsilon \rightarrow \infty$, we obtain

$$[I_\theta(\epsilon)]_{\epsilon \rightarrow \infty} \approx I_\theta^{(0,M)}(\epsilon) \left[1 + 4\pi An \left\{ \frac{1}{6} \frac{c^{1/3}}{\lambda} \epsilon^{1/6} - x^{-2} \left[1 + x \left(\frac{c}{\epsilon} \right)^{1/6} \right] \right\} \right. \\ \left. \times \exp \left[-x \left(\frac{c}{\epsilon} \right)^{1/6} \right] \right]_{\epsilon \rightarrow \infty} \rightarrow A \left(\frac{\epsilon}{c} \right)^{1/6} I_\theta^{(0,M)}(\epsilon), \left[\frac{I_\theta(\epsilon)}{I_\theta^{(0,M)}(\epsilon)} \right]_{\epsilon \rightarrow \infty} \rightarrow A \left(\frac{\epsilon}{c} \right)^{1/6} . \quad (20)$$

From (19) and (20) it follows that, if we take into account the correlational corrections to the ideal-gas expression for $I_\theta(\epsilon) = I_\theta^{(0,M)}(\epsilon)$, the behavior of the energy-loss spectrum gets complicated by interatomic correlations, and this complication has a statistical nature.

It is particularly interesting to stress that the relation of $I_\theta(\epsilon)$ to $I_\theta^{(0)}(\epsilon)$ asymptotically increases with increasing ϵ , as a power law $\sim \epsilon^{1/6}$ [see (20)] and, exponentially, rapidly tends to 1 with decreasing ϵ in accordance with (19). Let us look now at the spectral-line shape, particularly in the critical point of the phase transition. For this aim, we shall put the inverse correlational radius equal to zero. In this case, the first integral in large parentheses in (15) can be calculated with the help of change of variables and integration by parts; we obtain, as a result,

$$I_\theta(\epsilon) = I_\theta^{(0,M)}(\epsilon) \left[1 + 2\pi An \left\{ \left(\frac{c}{\epsilon} \right)^{1/3} + \Gamma(\frac{2}{3}) c^{1/3} (\pi\lambda^2)^{-3/4} \epsilon^{5/12} \exp \left[\frac{\pi\lambda^2}{2\epsilon} \right] \right. \right. \\ \left. \times \left[\frac{5}{6} W_{1/12, -1/4} \left(\frac{\pi\lambda^2}{\epsilon} \right) - \left(\frac{\pi\lambda^2}{\epsilon} \right)^{1/2} W_{7/12, -3/4} \left(\frac{\pi\lambda^2}{\epsilon} \right) \right] \right\} \right], \quad (21)$$

where $W_{\nu,\mu}(z)$ are the functions of Whittaker,¹⁰ $\Gamma(\frac{2}{3})$ is the Γ function. For the asymptotic value $I_\theta(\epsilon)$ in the formal limit $\epsilon \rightarrow 0$, taking into account the properties of $W_{\nu,\mu}(z)$;¹⁰

$$W_{\nu,\mu}(z) |_{z \rightarrow \infty} \approx e^{-z/2} z^\nu ,$$

we obtain

$$[I_\theta(\epsilon)]_{\epsilon \rightarrow 0+} \approx I_\theta^{(0,M)}(\epsilon) \left[1 + 2\pi An \left(\frac{c}{\epsilon} \right)^{1/3} \left\{ 1 + \Gamma \left(\frac{2}{3} \right) \left[\frac{5}{6} \left(\frac{\pi\lambda^2}{\epsilon} \right)^{-2/3} - \left(\frac{\pi\lambda^2}{\epsilon} \right)^{1/3} \right] \right\} \right] . \quad (22)$$

In the limit $\epsilon \rightarrow \infty$, using (21) and

$$W_{\nu,\mu}(z) |_{z \rightarrow 0} \approx \frac{\Gamma(-2\mu)}{\Gamma(\frac{1}{2} - \mu - \nu)} z^{\mu+1/2} e^{-z/2} , \quad (23)$$

we obtain

$$[I_\theta(\epsilon)]_{\epsilon \rightarrow \infty} \approx I_\theta^{(0,M)}(\epsilon) \left[1 + 2\pi An \left(\frac{c}{\epsilon} \right)^{1/3} + A \left(\frac{\epsilon}{c} \right)^{1/6} \right]_{\epsilon \rightarrow \infty} \rightarrow A \left(\frac{\epsilon}{c} \right)^{1/6} I_\theta^{(0,M)}(\epsilon) . \quad (24)$$

This result is in exact accordance with (20), which was obtained outside the critical point. Thus, from (20) and (24), it follows that the asymptotic behavior of the spectral line ($\epsilon \rightarrow \infty$) is modified due to interatomic correlations, but it is practically not influenced by the approach of the critical region.

On the contrary, the central part of the line, comparable to the Doppler width, is strongly dependent on the distance to the critical point [see (19) and (22)]. The relation of the cross section of energy loss $I_\theta(\epsilon)$ to the ideal-gas approximation of this function $I_\theta^{(0)}(\epsilon)$, in the critical point, changes in a formal limit $\epsilon \rightarrow 0$ as a power law, instead of the exponential decrease, in accordance with (19), which takes place outside the critical point.

Because the parameters ϵ , n , and c belong (see Sec. II) to fixed intervals of values, namely, $\epsilon \in [10^{-2}, 10^{-1}]$ eV, $n \in [10^{-2}, 10^{-3}]$ Å⁻³, and $c \in [10, 10^2]$ eV Å⁶, in practice, it is necessary to investigate the estimating of the behavior of CSR, taking into account these conditions. With parameters ϵ , n , and c already adopted, the dimensionless argument of Whittaker's functions in (21), $\pi\lambda^2/\epsilon$, belongs to the interval $\pi\lambda^2/\epsilon \in [10^{-3}, 1]$. Thus, in (21), outside the neighborhood of the resonances with width $\sim 10^{-2}$ eV, approximately, we can use series expansions (23) for Whittaker's functions $W_{v,\mu}(\pi\lambda^2/\epsilon)$, instead of the general expressions. This means that expression (24), in our model, approximately describes the behavior of CSR in the neighborhood of distinctive resonances $\epsilon \in [10^{-2}, \infty]$ eV, particularly in the critical point; and besides, the accuracy of this expression increases with increasing ϵ .

Thus, as follows from (19)–(24) in the behavior of the cross section of energy loss near the distinctive resonances $I_\theta(\epsilon)$, in the wide interval of thermodynamic parameters, scaling takes place in relation to the ideal-gas limit of this function $I_\theta^{(0)}(\epsilon)$, and the value $\pi\lambda^2/\epsilon$ plays a role in the scaling argument.

Expression (19)–(24) are a good test for the investigation of the influence of many-particle correlational effects in the theory of atomic line shapes in dense disordered systems. Moreover, this theory, practically without any change, can be applied to the qualitative and quantitative description of the atomic line shape in the absorption and emission⁷ of radiation in a similar system. (See *Note added in proof*.) These questions will be investigated in our next papers.

VI. FRACTON INTERPRETATION OF THE SCALING BEHAVIOR OF CSR

It is necessary to stress that the expressions (19)–(24) show that, in our model, in the phenomena of inelastic charged-particle scattering in dense disordered systems, scaling takes place with reference to the analogous scattering in rare (or ideal) disordered systems.

The results that we obtained in Sec. V can be interpreted in terms of fractals which are used for geometrical interpretation of self-similar phenomena in the wide range of disordered systems (see, for example, Ref. 12). In connection with this, it should be pointed out that the rela-

tion $I_\theta(\epsilon)/I_\theta^{(0)}(\epsilon) \equiv N(\epsilon)$, which was described in Sec. V, reproduces the normalized density of state [which was determined by the value of $\mu(R)$].

In our model, many-particle fluctuational clusters, which include an excited molecule and were introduced when scattering occurs by means of correlational expansions, play a role in fractal objects. Fractal symmetry, as opposed to Euclidean symmetry, requires three dimensionalities to contain a physical description of the excitations of a fractal network; d is the Euclidean dimension, D is the fractal dimension, and \bar{d} is the spectral dimension (or fracton). The concept of fractons was first introduced in the context of the study of the dynamics of self-similar fractal objects.¹³ In accordance with the central idea, let us assume that the density of state $N(\epsilon)$ is described with the help of such a relation,

$$N(\epsilon) \sim \epsilon^{\bar{d}-1}. \quad (25)$$

From a comparison of (25) and (19)–(24), it follows that, in the asymptotic region of single resonance in the energy-loss spectrum (or the atomic line in absorption or emission) for the model of localized excitation to the critical region, the fracton dimension is equal to $\bar{d} = \frac{7}{6}$.

It is interesting to compare the fracton dimension of our model $\frac{7}{6} = 1.16$ with the value $\bar{d} = \frac{4}{3} = 1.33$, which was obtained in the task of percolation.¹⁴ These results are in good accordance with the Anderson localization scaling theory, in which it was shown that, for $\bar{d} < 2$, the fracton eigenstates are localized.¹⁴ Thus localization can occur by virtue of the geometrical connectivity properties of the atomic network, in which case we really obtain the possibility of describing localized excitations in many-particle systems as fractons. The fracton's regime, in our model, takes place starting from the small neighborhood of distinctive resonance $\epsilon \sim 10^{-2}$ eV, and formally with universal dimension last to $\epsilon \rightarrow \infty$, which reflects the fact that due to localization of excitation, the main part of the resonance formed by short-range interatomic correlations (which are insusceptible to the critical behavior). In the small neighborhood of resonances with losses $\Delta < \epsilon < 10^{-2}$, which are formed by long-range interatomic correlations, an essential change of line shape takes place, particularly in the critical point (see Sec. V). This shows that the geometry of the fractal, even if determinant, can induce localization. It is believed that in real random fractals, the fractons are always localized excitations.

VII. DIRECT DETERMINATION OF THE RADIAL DISTRIBUTION FUNCTION BY INELASTIC ELECTRON SCATTERING

In the system with moderate density, we can use the finite length of the correlation series (2). For example, if we take into account only the first two terms of the correlational series (2), we obtain the expression that describes the cross section of the energy loss by scattering on a system which consists of “two-atom quasimolecules,” of which the regular part is equal to

$$I_\theta(\epsilon) = \frac{1}{2} \left(\frac{e}{E\theta^2} \right)^2 n^2 \int g_2(R) \sum_{(\alpha)} Z_{\alpha,2}(\mathbf{q}, \mathbf{R}) \times \delta(\epsilon + \mu_{\alpha,2}(\mathbf{R})) d\mathbf{R}. \quad (26)$$

In the quasimolecular form, we find two equal atoms, one of which is in an excited state; the electronic states are divided into even (g) and odd (u), in accordance with the property of the wave functions; they conserve or change the sign under the inversion of electronic variables in the plane of symmetry. Thus, α in (26) may be equal to g or u . In the zeroth-order perturbation theory, on interatomic interactions, when overlap effects are neglected, the wavefunctions of the ground state $-\Psi_0(\{\mathbf{r}\}, \mathbf{R})$ and the excited states $-\Psi_{1,\alpha}(\{\mathbf{r}\}, \mathbf{R})$ of the two-atom complex in which both atoms shift from one to the other on vector R , are expressed by the wave functions corresponding to the ground state $\Psi_0([\mathbf{r}_1])$ and the excited state $\Psi_1([\mathbf{r}_2 - \mathbf{R}])$ of isolated atoms, with the help of the following rule:

$$\begin{aligned} \Psi_0(\{\mathbf{r}\}, \mathbf{R}) &= \psi_0([\mathbf{r}_1])\psi_0([\mathbf{r}_2 - \mathbf{R}]), \\ \Psi_{1,\alpha}(\{\mathbf{r}\}, \mathbf{R}) &= 2^{-1/2} \{ \psi_0([\mathbf{r}_1])\psi_1([\mathbf{r}_2 - \mathbf{R}]) \\ &\quad \pm \psi_1([\mathbf{r}_1])\psi_0([\mathbf{r}_2 - \mathbf{R}]) \}, \end{aligned} \quad (27)$$

where $\{\mathbf{r}\}$ is the set of electronic variables of the pair of atoms, $[\mathbf{r}_1]$ and $[\mathbf{r}_2 - \mathbf{R}]$ are, respectively, the set of electronic coordinates of the first and second atom and the \pm sign corresponds to $\alpha=g$ and u .

With the help of the determination of the Fourier transform of the charge-density operator in the two-atom complexes, namely,

$$\begin{aligned} \rho_{10,\alpha}^{(2)}(\mathbf{q}, \mathbf{R}) &= \sum_e \int \cdots \int e^{-i\cdot\mathbf{q}\cdot\mathbf{r}_e} \Psi_{1,\alpha}^*(\{\mathbf{r}\}, \mathbf{R}) \\ &\quad \times \Psi_0(\{\mathbf{r}\}, \mathbf{R}) \{d\mathbf{r}\}, \end{aligned} \quad (28)$$

and expression (27), we obtain

$$Z_{\alpha,2}(\mathbf{q}, \mathbf{R}) = 2Z_1(\mathbf{q}) \times \begin{cases} \cos^2(\mathbf{q}\cdot\mathbf{R}/2), & \alpha=g \\ \sin^2(\mathbf{q}\cdot\mathbf{R}/2), & \alpha=u \end{cases} \quad (29)$$

In accordance with the determination, $\mu_{\alpha,2}(R)$ is equal to $E_{10}^{(\alpha)}(R) - E_{00}(R)$, where $E_{10}^{(\alpha)}(R)$ and $E_{00}(R)$ are the energies of the interatomic interactions in the two-particle fluctuational complexes, respectively, in the excited and the ground states, which will be considered in the adiabatic approximation, for example, with the help of perturbation theory.

It is necessary to distinguish the cases when the excited states are dipole-forbidden or dipole permitted, i.e., when,

for the correspondent (crossing) transition $0 \rightarrow 1$, the matrix element from the operator of the dipole moment of the molecule \mathbf{d}_{10} equals zero or not. In the dipole approximation, the interatomic interaction is described with the help of such an operator, \hat{V} ,

$$\hat{V} = \frac{(\mathbf{d}_1 \cdot \mathbf{d}_2) - 3(\mathbf{d}_1 \cdot \mathbf{n})(\mathbf{d}_2 \cdot \mathbf{n})}{R^3}, \quad (30)$$

which is called the operator of the dipole-dipole interaction. Here \mathbf{d}_1 and \mathbf{d}_2 are the dipole moments of both atoms, and \mathbf{n} is the unit vector in the direction in which the centers of charge of the atoms are connected.

Let us consider, for example, that the ground states and the excited states are, respectively, $1S$ and $1P$ states (i.e., the transition $0 \rightarrow 1$ is dipole permitted). The matrix elements of the operator \hat{V} are given by

$$\begin{aligned} V_{1i}^{(\alpha)}(\mathbf{R}) &\equiv \langle \Psi_{1i,\alpha} | \hat{V} | \Psi_{1i,\alpha} \rangle \\ &= \int \cdots \int \Psi_{1i,\alpha}^*(\{\mathbf{r}_j\}, \mathbf{R}) \hat{V} \Psi_{1i,\alpha}(\{\mathbf{r}_j\}, \mathbf{R}) \{d\mathbf{r}_j\}, \end{aligned} \quad (31)$$

which may be calculated by using the wave functions (27), namely,

$$V_{1i}^{(\alpha)}(\mathbf{R}) = \pi_{ia} \frac{d_{10}^2}{R^3}, \quad (32)$$

where

$$\pi_{ia} = \begin{cases} -2, & \alpha=g, \quad i=z \\ 1, & \alpha=g, \quad i=x,y \\ 2, & \alpha=u, \quad i=z \\ -1, & \alpha=u, \quad i=x,y. \end{cases}$$

Here $i=x, y, z$ means three possible reciprocally perpendicular directions of orientation of the angular moment of the excited molecule; besides, $i=z$ if the angular moment and the axis of the quasimolecule are collinear. Finally, with the help of (26) and (32), we obtain the following expression for $I_\theta(\epsilon)$ in this case:

$$\begin{aligned} I_\theta(\epsilon) &= \frac{2\pi}{3} a'(\theta) \frac{d_{10}^2}{\epsilon^2} Z_1(\mathbf{q}) n^2 \\ &\quad \times \sum_{s=-2}^2 \left[1 + \text{sgn}(s) \frac{\sin(qR_s)}{qR_s} \right] \theta(R_s) g_2(R_s), \end{aligned} \quad (33)$$

where

$$\theta(x) = \begin{cases} 1, & x > 0 \\ 0, & x < 0 \end{cases}, \quad R_s = \left[-s \frac{d_{10}^2}{\epsilon} \right]^{1/3}. \quad (34)$$

If we consider the dipole-forbidden transition, for example, the $1S \rightarrow 2S$ transition, as a function of the shift not equal to zero only in the second order of the perturbation theory on interatomic interactions and dependent only from the module of the interatomic distance R , then $\mu_{\alpha,2}(R) \approx -(C_\alpha/R^6)$ (where C_α is the atomic constant). In this case, we obtain

$$I_\theta(\epsilon) = \frac{1}{4} a(\theta) Z_1(q) n^2 \left[\sum_v \frac{R_v^2 \left[1 + \frac{\sin(qR_v)}{qR_v} \right]}{\left| \frac{d\mu_{g,2}(R)}{dR} \right|_{R=R_v(\epsilon)}} g_2(R_v(\epsilon)) + \sum_{v'} \frac{R_{v'}^2 \left[1 - \frac{\sin(qR_{v'})}{qR_{v'}} \right]}{\left| \frac{d\mu_{u,2}(R)}{dR} \right|_{R=R_{v'}(\epsilon)}} g_2(R_{v'}(\epsilon)) \right], \quad (35)$$

where R_v and $R_{v'}$, respectively, are the solutions of the equations

$$\begin{aligned} \mu_{g,2}(R) &= \hbar(\omega - \omega_1) = -\epsilon, \\ \mu_{u,2}(R) &= \hbar(\omega - \omega_1) = -\epsilon, \\ \mu_{a,2}(R) &= -C_a/R^6. \end{aligned} \quad (36)$$

In the general case, if, for any frequency $\bar{\omega}$, one of the equations in (36) has a divisible root, then in the limit $\omega \rightarrow \bar{\omega}$ function, $I_\theta(\epsilon)$ will be sharply increased. Near the similar frequencies in the energy-loss spectrum, sharp peaks may appear. These singularities are analogous to the singularities of van Hove for the spectral densities of state. Let us consider that, for such a frequency interval, there exists a solution only for one of the equations in (36). Then, for the corresponding interval of the distances, we can directly determine the value of $g_2(R(\omega))$ by using Eqs. (35) and (36). If the splitting of the distinguishing level is negligible,

$$|\mu_{g,2}(R) - \mu_{u,2}(R)| \sim \Delta \ll |\mu_{g,2}(R) + \mu_{u,2}(R)|/2, \quad (37)$$

we obtain a more simple form of expression (35), namely,

$$\begin{aligned} I_\theta(\epsilon) &= \frac{1}{2} a(\theta) n^2 Z_1(q) R^2 \left| \frac{dR}{d\epsilon} \right| g_2(R(\epsilon)), \\ \mu_{a,2}(R) &= \mu(R(\epsilon)) = -\epsilon = -c/R^6. \end{aligned} \quad (38)$$

Expression (38) can be rewritten in the following form:

$$I_\theta(\epsilon) = \frac{c}{12} a(\theta) Z_1(q) \epsilon^{-3/2} n^2 g_2 \left[\left(\frac{c}{\epsilon} \right)^{1/2} \right], \quad (39)$$

from which, in the ideal-gas limit, when $g_2 = 1$, follows the famous result, $I_\theta(\epsilon) \sim \epsilon^{-3/2}$.⁴ Expressions (33)–(38), in principle, make possible a direct determination of $g_2(R)$ in the individual points in simple disordered systems, instead of the usual procedure, which includes the inverse Fourier transform of the structure factors of scattering [due to irregular convergence of the Fourier series, this procedure is a source of inexactitudes in the determination of $g_2(R)$ (Ref. 14)].

VIII. CONCLUSIONS

Thus, in addition to the conclusions of Ref. 1, the main results are the following.

(1) Within the framework of some approximations (Born approximation, adiabaticity, localized excitation) which are adequate for a deterministic construction of our task and which also correspond to conditions of possible experiments, we proved the possibility of complete summing of the correlation expansion for the cross section of energy losses $I_\theta(\epsilon)$ in EELS of the statistically determined simple disordered systems for any approximation of the many-particle distribution function which is determined beforehand.

(2) On the basis of (1), it was shown that near the critical point the normalized “spectral density of states” (in EELS) in the small neighborhood of distinctive narrow dipole-forbidden long-living resonance in the limit when $\epsilon \rightarrow 0 + \Delta$ is represented by a power law instead of an exponential law, which takes place outside of the critical region. Thus the small neighborhood of narrow resonance in EELS close to the critical point displays the scaling behavior relative to the corresponding “gas” limit.

(3) In systems with moderate densities, by using higher-resolution EELS we can directly (without integral transformation) determine the radial distribution function $g_2(R)$ in a fixed number of points by special construction of the deterministic conditions of the experiment.

Finally, we note that typical examples of disordered systems which were investigated *vide supra* are the inert gases and liquids.

Note added in proof. After submitting the manuscript the authors obtained information about the work of S. Mukamel, P. S. Stern, and D. Ronis in *Spectral Line Shapes*, edited by K. Burnett (de Gruyter, Berlin, 1983), Vol. 2, in which the behavior of the optical line shape near the critical point was investigated in terms of so-called line-shape functions. Within the framework of the approach in which the evaluation of the line-shape function was reduced to the calculation of the three-particle, two-time correlation function, it was shown that the critical behavior of this correlation function is being probed by the optical-broadening experiment.

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