## The role of surface scattering of electrons in the excitation of current states in metals

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The effect of surface scattering of electrons on the current states in metals has been studied theoretically. It is found that the parameter that determines the current state is not the same as the magnetodynamic nonlinearity parameter and it depends on the state of the sample surface. Improving the specular quality of the surface shifts the current state hysteresis towards larger radio wave amplitudes. It is shown that hysteresis loops in the magnetic moment of the sample exist in a limited range of amplitudes, but for pure specular reflection vanish completely. When surface scattering is taken into account it is possible to explain the frequency independence of the current states.

In recent years there have been a large number of experimental and theoretical investigations of the magnetodynamic nonlinearity in pure metals (see, e.g., Refs. 1-11). Among the more conspicuous nonlinear responses to electromagnetic excitation are the current states. The macroscopic manifestation of the magnetodynamic nonlinearity in current states is the formation of a magnetic moment in a sample that is irradiated by large-amplitude radio waves. According to Ref. 5, the nonlinearity mechanism that brings about current rectification is associated with the fact that the conduction electron trajectories are determined by a nonuniform magnetic field which is the sum of the external field  $\mathbf{h}_0$ and the magnetic field  $\mathbf{H}(x,t)$  that is induced in the metal by the incident wave (the x axis points down into the metal). As long as the total magnetic field is pointing in one direction everywhere, the bulk electrons move in trajectories that are close to Larmor trajectories (see Fig. 1a). However, when the field points in both directions, then in place of the Larmor electrons there is a group of trapped electrons (see Fig. 1b). their trajectories oscillate around the plane  $x = x_0(t)$ , on which

$$H(x_0, t) + h_0 = 0$$
 (**H**(x, t) ||**h**\_0).

Since the total magnetic field governs the shapes of the trajectories of the effective electrons, it also governs the time dependence of the conductivity of the metal. As a result a rectified current is induced in the sample, and associated with this is a constant nonuniform magnetic field h(x). The field h(x), which vanishes at the surface of the metal varies within a distance on the order of the skin depth and assumes the value  $h(\infty)$  in the interior of the sample. When the amplitude  $\mathcal{H}$  of the incident wave is greater than some threshold value  $\mathcal{H}_{cr}$ , the dependence of the induced field  $h(\infty)$  on the external field  $h_0$  will exhibit hysteresis and discontinuous changes in the magnetic moment of the sample occur as  $h_0$  is changed. This sort of behavior of the magnetic moment has been observed experimentally in bismuth<sup>6,7</sup> and later was studied in detail in a number of metals.<sup>1,8,9</sup> A theory of current states has been formulated<sup>10,11</sup> that is consistent with

the main experimental data.

This paper consists of a further development of the theory of current states, in which the role of surface scattering of electrons is investigated. Until now the effect of surface quality on the current states has not been considered. In part the reason for ignoring surface effects is that the group of electrons that are responsible for the current states do not interact at all with the surface. However, in this article we show that surface scattering plays an extremely important role. For instance, in the case of nearly specular reflection of electrons from a metal surface the conductivity of the grazing electrons can be comparable to that of the trapped electrons or even exceed it. Since the group of grazing electrons is always present (see Fig. 1a and 1b) any abrupt changes in the conductivity due to the appearance and disappearance of the group of captured electrons will be smoothed out. When the quality of the surface is improved, the conductivity of the grazing electrons begins to dominate over that of the trapped electrons, and there is no hysteresis in the current states no matter how great the amplitude of the incident wave is. This occurs (as shown below) in those cases where the bulk relaxation frequency is greater than the surface relaxation frequency.

Taking into account the interaction of the electrons with the surface allows a natural interpretation of some experimental facts that have no explanation within the framework of the theory of Ref. 11. For example, the dependence of the threshold value  $\mathcal{H}_{cr}$  on the frequency  $\omega$  of the incident wave,  $\mathcal{H}_{cr} \propto \omega^{-1/2}$ , as follows from the theory,<sup>11</sup> is observed only for tin. Furthermore, the magnitude of the induced field in zero external magnetic field (according to Ref. 11) for incident wave amplitudes  $\mathcal{H}$  attainable in present-day experiments should, to a high degree of accuracy, be equal to  $4\mathcal{H}/\pi$ . However, in these experiments this degree of hysteresis is not observed and the loops that in fact occur are far removed from the limiting one predicted by the theory of current states.<sup>11</sup> This fact and the independence of  $\mathcal{H}_{cr}$  on  $\omega$  observed in the majority of the experiments can be explained within a theory of current states that takes into

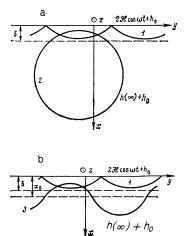


FIG. 1. Trajectories of effective electrons in a total magnetic field of constant sign (a) and alternating sign (b). 1) grazing electrons; 2) Larmor electrons; 3) trapped electrons.

account surface scattering of the electrons.

An indication that the surface quality has an effect on the parameters that characterize the current states can be found in the experimental investigation of Ref. 9. Unfortunately, that article does not say just what this effect is. Because the appropriate information is insufficient, we are not able to make a comparison of the theory with the experiment of Ref. 9.

1. Let us consider a wave of amplitude  $\mathcal{H}$  and frequency  $\omega$  incident on a semiinfinite metal situated in a constant and uniform magnetic field  $\mathbf{h}_0$  which is parallel to the metal surface. The wave is polarized so that its magnetic field is collinear with the vector  $\mathbf{h}_0$ . We shall take the *x* axis pointing down into the metal perpendicular to the surface and the *z* axis parallel to  $\mathbf{h}_0$ . The amplitude and frequency of the incident wave are such as to satisfy the conditions of the quasistatic ( $\omega \ll \nu$ ) anomalous skin effect:

$$\delta \ll l, R = c p_F / 2e \mathcal{H}. \tag{1}$$

Here  $\delta$  is the skin depth, c is the velocity of light,  $e, p_F, v$ , and l, are, respectively, the charge, Fermi momentum, bulk relaxation frequency, and mean free path of the electrons.

The electromagnetic field in the metal

$$\mathbf{E}(x,t) = \{0, E(x,t), 0\}, \quad \mathbf{H}(x,t) = \{0, 0, H(x,t)\} \quad (2)$$

is determined by the Maxwell equations

$$-\frac{\partial H(x,t)}{\partial x} = \frac{4\pi}{c} j(x,t), \quad \frac{\partial E(x,t)}{\partial x} = -\frac{1}{c} \frac{\partial H(x,t)}{\partial t} \quad (3)$$

with boundary conditions which to the accuracy of the impedance can be written in the form

$$H(0,t) = 2\mathcal{H}\cos\omega t, \quad H(\infty,t) = h(\infty).$$
(4)

2. In order to put Eq. (3) into closed form, it is necessary to find the relation between the current density j(x,t)and the electromagnetic field in the metal. For this purpose we use the ineffectiveness concept, which has been extended to nonlinear situations<sup>11</sup> and which, to within a real factor of order unity, gives the right results. The conductivities of the Larmor, the trapped and the surface electrons, to which we shall subsequently apply the nonlinear concept of ineffectiveness, are obtained in exactly the same way as in Refs. 11 and 12, and they have the form

$$\sigma_{L} = \sigma_{0} \frac{\delta}{l} \operatorname{cth} v T_{L}, \quad \sigma_{t} = \sigma_{0} \frac{\delta}{l} \operatorname{cth} v T_{t},$$

$$\sigma_{s} = \sigma_{0} \frac{\delta}{l} \frac{1 + \rho \exp(-2vT_{s})}{1 - \rho \exp(-2vT_{s})},$$
(5)

where  $\sigma_0$  is the static conductivity of the infinite metal,  $\rho$  is the probability of specular reflection of an electron at the boundary, and  $2T_L$ ,  $2T_s$ , and  $2T_t$  are the characteristic periods of the Larmor, the surface, and the trapped electrons:

$$2T_s = \frac{b}{v}, \quad 2T_t = \frac{b}{v} \frac{2\mathscr{H}}{|h_0 + h(\infty)|}, \quad b = \frac{(8R\delta)^{\frac{1}{b}}}{l}.$$

Here we introduce the nonlinearity parameter b, which characterizes the contribution of the trapped electrons to the conductivity (see Ref. 11).

The structure of expressions (5) is quite transparent. The spatial dispersion is taken into account by the factor  $\delta/l$ , as is usual for anomalous skin conductivities. It reflects the fact that not all the effective electrons, but only those that are incident on the skin layer, interact with the electromagnetic field. The factors containing the periods of the motion taken into account for the Larmor and the trapped electrons the probability of multiple returns to the skin layer, and for the surface electrons take into account the probability of multiple returns to the skin layer, and for the surface electrons take into account the probability of multiple reflection from the metal boundary. The specularity parameter  $\rho$  which enters into (5) depends strongly on the angle at which the grazing electrons encounter the metal boundary.

At those instants of time when the magnetic field points in the same direction everywhere,

$$\frac{2\mathscr{H}\cos\omega t + h_0}{h(\infty) + h_0} > 0, \tag{6}$$

only Larmor and surface electrons are present in the metal (see Fig. 1a). However, when the inequality (6) is not satisfied, the magnetic field alternates in direction. In these intervals of time the conductivity of the metal is determined by the trapped and the grazing electrons (Fig. 1b).

For simplicity we shall consider the case  $2\nu T_L \ge 1$ , i.e., we shall neglect the return of the Larmor electrons to the skin layer. In this case the total time-dependent conductivity of all the groups of electrons can be written in the form

$$\sigma(t) = \frac{2\sigma_0}{1 - \rho \exp(-b)} \frac{\delta}{l} S(\varphi), \qquad (7)$$

where

$$\begin{split} \varphi &= \omega t, \quad S(\varphi) = \theta_{+} + \alpha(\varphi) \theta_{-}, \\ \alpha &= \frac{1 - \rho \exp\left[-b\left(1 + |\overline{x}|^{-1}\right)\right]}{1 - \exp\left(-b\left|\overline{x}\right|^{-1}\right)}, \\ \theta_{\pm} &= \theta \left(\pm \frac{2\mathcal{H}\cos\varphi + h_{0}}{h(\infty) + h_{0}}\right), \quad \overline{x} = \frac{h_{0} + h(\infty)}{2\mathcal{H}}, \end{split}$$

and  $\theta(x)$  is the Heaviside function, equal to zero for x < 0

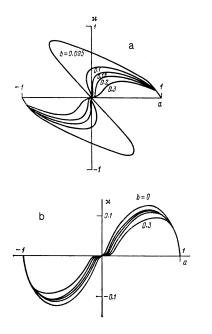


FIG. 2. Dynamics of the variation of  $\kappa(a)$  with increasing wave amplitude (with decreasing parameter b) for diffuse (a) and specular (b) reflection of electrons from the surface.

and unity for x > 0.

3. The solution of Maxwell's equations within the ineffectiveness scheme leads to an equation for the induced field:

$$\kappa = \frac{(1-a^2)^{\frac{1}{b}}\operatorname{sign}\overline{\varkappa}}{\pi[\exp(b/|\overline{\varkappa}|)-1][1-\rho\exp(-b)]^{-1}+\arccos(-a\operatorname{sign}\overline{\varkappa})}$$

$$\kappa = h(\infty)/2\mathcal{H}, \quad a = h_0/2\mathcal{H}.$$
(8)

The skin depth  $\delta$  is given by the formula

$$\delta = \left[\frac{c^2 l}{8\pi\omega\sigma_0}\mu(1-\rho\exp(-b))\right]^{\eta_0},\tag{9}$$

where

$$\mu = \frac{1}{2\pi} \oint \frac{d\varphi}{S(\varphi)}.$$

We note that in the special case of diffuse reflection  $(\rho = 0)$  Eq. (8) describes the family of curves  $\varkappa(a)$  obtained in Ref. 11 (see Fig. 2a). The different curves of this family correspond to different values of the nonlinearity parameter b. The curves of  $\varkappa(a)$  corresponding to a nonlinearity parameter smaller than a certain value  $b_{\rm cr}^{\rm dif} \approx 0.2$  have an S-shape in the a- $\varkappa$  plane. This means that for  $b = b_{\rm cr}^{\rm dif}$  hysteresis loops are produced in the magnetic moment of the sample as a function of the external magnetic field  $\mathbf{h}_{0}$ .

The criterion for the appearance of hysteresis loops proves to be quite different in the case of nondiffuse electron scattering from the metal surface. It may be established by noting that the curve  $\kappa(a)$  immediately preceding the Sshaped curves has two centrosymmetric points ( $\pm a_{\rm cr}$ ,  $\pm \kappa_{\rm cr} \equiv \kappa(a_{\rm cr})$ ) with vertical slopes. The two equations  $\partial a/\partial x = \partial^2 a/\partial x^2 = 0$  along with Eq. (8) constitute a system of equations that enable us to find  $a_{\rm cr}$ ,  $\kappa_{\rm cr}$ , and the value of the nonlinearity parameter  $b_{\rm cr} \equiv b(\mathscr{H}_{\rm cr})$  at which the hysteresis in induced:

$$\frac{\pi}{1-\rho\exp\left(-b\right)}\left[\exp\left(\frac{b}{|\overline{x}|}\right)-1\right] = \frac{\operatorname{sign}\overline{\varkappa}}{\varkappa},$$
$$\frac{\pi b}{1-\rho\exp\left(-b\right)}\exp\left(\frac{b}{|\overline{\varkappa}|}\right) = \left(\frac{\overline{\varkappa}}{\varkappa}\right)^{2}, \quad (10)$$
$$\frac{\pi b}{1-\rho\exp\left(-b\right)}\left(1+\frac{b}{2|\overline{\varkappa}|}\right)\exp\left(\frac{b}{|\overline{\varkappa}|}\right) = \left(\frac{\overline{\varkappa}}{\varkappa}\right)^{3}.$$

Here we have expanded in the small quantities  $a_{\rm cr}$  and  $\varkappa_{\rm cr}$ . As an analysis of Eq. (8) shows, they are numerically small in the diffuse case  $(|a_{\rm cr}^{\rm dif}|, |\varkappa_{\rm cr}^{\rm dif}| \leq 1)$  while in the case of nearly specular reflection there is an additional small parameter  $1 - \rho$ . An analytic solution of system (10) is not possible. However, in the new variables

$$a' = \frac{a}{1 - \rho \exp(-b)}, \quad \varkappa' = \frac{\varkappa}{1 - \rho \exp(-b)},$$
$$B = \frac{b}{1 - \rho \exp(-b)}, \quad (11)$$

the system (10) takes on the form that it had in the old variables with  $\rho = 0$ . This allows us to relate the general solution  $(0 \le \rho \le 1)$  of system (10) with its special solution  $(\rho = 0)$ . An indeterminacy appears at the point

$$a_{\rm cr} = a_{\rm cr}^{\rm dif} b_{\rm cr} / b_{\rm cr}^{\rm dif}, \quad \varkappa_{\rm cr} = \varkappa_{\rm cr}^{\rm dif} b_{\rm cr} / b_{\rm cr}^{\rm dif}, \quad (12)$$

where the parameter B is equal to

$$B(\mathscr{H}_{\rm cr}) = b_{\rm cr}^{\rm dif} \approx 0.2.$$
(13)

Equation (13) determines the threshold value of the amplitude of the incident wave for an arbitrary type of reflection of the electrons from the metal surface. From this equation can be seen the importance of the role played by the parameter  $B(\mathcal{H})$  that we have introduced. In the case of an arbitrary reflection  $(0 \le \rho \le 1)$  the parameter  $B(\mathcal{H})$  plays the same role for the dynamics of loops that the nonlinearity parameter *b* plays for the dynamics of diffuse loops  $(\rho = 1)$ . The presence or absence of current state hysteresis depends on the value of  $B(\mathcal{H})$ . Moreover, a detailed analysis of Eq. (8) in various regions of *a* and  $\varkappa$  shows that the shape of the curves  $\varkappa(a)$  and their characteristic sizes are governed by the parameter  $B(\mathcal{H})$ .

From (11) and (13) it can be seen that a deviation from diffuse reflection shifts the threshold value of  $\mathscr{H}_{cr}$  towards larger amplitudes and decreases the amount of the hysteresis. In the case of partially diffuse reflection  $(1 - \rho \sim 1)$  the dynamics of the curves  $\varkappa(a)$ , being determined by the value of  $B \sim b/(1-\rho) \sim b$ , differs only insignificantly from that of diffuse loops.

A rather different situation occurs in the case of nearly specular reflection. From the structure of the parameter  $B(\mathcal{H})$  it can be seen that the onset of hysteresis and the appearance of an appreciable induced field  $(|h(\infty)| \sim 2\mathcal{H})$ are shifted towards very large amplitudes, where  $b \ll b_{cr}^{dif}$ . For strong nonlinearity  $(b \ll 1)$  the parameter  $B(\mathcal{H})$  can be written in the form

$$B(\mathscr{H}) = \left(1 + \frac{1-\rho}{b}\right)^{-1}.$$
(14)

In the case of pure specular reflection ( $\rho = 1$ ) hysteresis in  $\kappa(a)$  is not possible for any amplitude  $\mathcal{H}$ . The dynam-

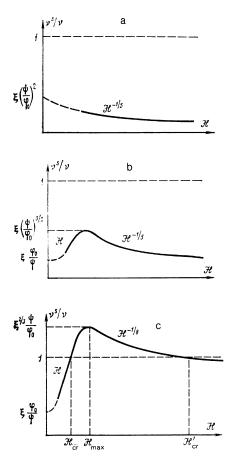


FIG. 3. Surface relaxation frequency of grazing electrons as a function wave amplitude.

ics of the variation of  $\kappa(a)$  with increasing amplitude  $\mathcal{H}$  for  $\rho = 1$  is shown in Fig. 2b.

4. In order to exhibit the physical meaning of the parameter  $B(\mathcal{H})$  we introduce, following Refs. 13 and 14, the effective surface relaxation frequency

 $v^s = (1-\rho)/2T_n$ 

and rewrite (14) in the form

$$B(\mathcal{H}) = (1 + v^{s}/v)^{-1}.$$
 (15)

From (15) it can be seen that the parameter  $B(\mathcal{H})$  and, consequently, the dynamics of the loops, their shape and characteristic sizes, are determined by the competing bulk and surface scattering of grazing electrons. For those values of  $\mathcal{H}$  for which bulk electron relaxation dominates the surface relaxation ( $v^{s} \ll v$ ), Eq. (13) has no solution. Consequently there is no current state hysteresis and the induced field is small ( $|h(\infty)| \ll 2\mathcal{H}$ ). Hysteresis arises when the surface and bulk relaxation frequencies are of the same order of magnitude.

$$\mathbf{v}^{\mathbf{s}}(\mathcal{H}_{\mathrm{cr}})/\mathbf{v} = (b_{\mathrm{cr}}^{\mathrm{dif}})^{-\mathbf{i}} - 1.$$
(16)

For amplitudes  $v^{s}(\mathcal{H}) \ge v$  hysteresis is well developed and is characterized by the value of the parameter

$$B(\mathcal{H}) = v/v^{s}(\mathcal{H}).$$
(17)

In order to analyze the dynamics of hysteresis loops as the amplitude  $\mathscr{H}$  is varied we shall use the results of Refs. 13 and 14, in which the authors obtained the dependence of the frequency  $v^s$  on the angle of incidence  $\varphi$  of the grazing electrons on the surface (in our case  $\varphi = (\delta/R)^{1/2}$ ). The fact that the reflection of the electrons from the surface is nearly specular imposes certain limits on the geometric size of the surface irregularities (see Refs. 13 and 14):

$$(\sigma/L)^2 \ll 1, \quad \xi \equiv \sigma^2 p_F / \hbar L \ll 1, \tag{18}$$

where  $\hbar$  is Planck's constant,  $\sigma$  is the rms height of the irregularities, and L is their mean lateral dimension. The first of the inequalities (18) means that the angle of inclination of the irregularities must be small and the second allows us to neglect diffraction for single scattering of an electron.

Let us consider the most typical situation of large-scale irregularities  $(\psi^2 \equiv 2\pi\hbar/p_F L \leq 1)$ , where  $\psi$  is the angular width of the scattering indicatrix). As the analysis shows, the possibility of obtaining current state hysteresis is determined by the relation between two parameters: the parameter  $\xi^{2/3}\psi$ , which depends on the surface quality, and the parameter  $\varphi_0 = \delta_0/1$  [where  $\delta_0 = (c^2 1/4\pi\omega\sigma_0)^{1/3}$ ], which characterizes the electrodynamic properties of the metal. The quantity  $\varphi_0$  is the angle of incidence of the effective electrons on the surface in the absence of magnetic fields.

If the surface is close to ideal, i.e., if  $\xi^{2/3}\psi < \varphi_0$  or

$$\frac{\delta_0}{l} > \left(\frac{\sigma^2 p_F}{\hbar L}\right)^{1/\epsilon} \frac{\sigma}{L},\tag{19}$$

then the surface relaxation frequency in any magnetic field is less than the bulk relaxation frequency (Fig. 3a and 3b). This means that the conductivity of the gliding electrons is greater than that of the trapped electrons and there is no current state hysteresis (as in the case of pure specular reflection). We note that condition (19) can be satisfied by decreasing the frequency of the incident wave or increasing the sample temperature T.

In order for current state hysteresis to be possible, the opposite limit from that defined by inequality (19) must be satisfied. In this case there is a range of fields for which the surface relaxation frequency is substantially greater than the bulk relaxation frequency (see Fig. 3b). Since the surface relaxation frequency is a nonmonotonic function of the magnetic field, hysteresis loops exist over a limited range of incident wave amplitude  $\mathcal{H}$ .

The onset of the current state hysteresis occurs when  $\mathcal{H}_{cr}$  reaches the threshold value

$$\mathscr{H}_{cr} \approx \frac{cp_F}{el} \left(\frac{2\pi\hbar}{p_F L}\right)^{\prime/2} \left(\frac{\sigma^2 p_F}{\hbar L}\right)^{-1} , \qquad (20)$$

which is independent of the frequency  $\omega$  and is determined by the sample temperature  $[\mathscr{H}_{cr} \propto \nu(T)]$  and the statistical characteristics of the surface. Further increase in the amplitude causes the hysteresis loops  $\varkappa(a)$  which are expanding rapidly,  $(B \approx \psi R / \xi \ 1 \propto \mathscr{H}^{-1})$ , to reach a maximum  $(B_{\min} = \varphi_0 / \xi^{2/3} \psi \leqslant 1)$  at the amplitude

$$H_{max} \approx \frac{cp_F}{e\delta_0} \frac{2\pi\hbar}{p_F L} \left(\frac{\sigma^2 p_F}{\hbar L}\right)^{-1/3} \, \infty \, \omega^{1/3}. \tag{21}$$

Then, with increasing amplitude, they very slowly get

smaller

$$(B \approx (\xi \psi^2)^{5/s} \overline{\varphi_0^{-1/s}} \ (R/l)^{-1/s} \infty \mathscr{H}^{1/s})$$

and vanish when the amplitude of the wave becomes

$$\mathscr{H}_{\rm cr}' \approx \frac{cp_F}{el} \left(\frac{\delta_{\bullet}}{l}\right)^{-9} \left(\frac{2\pi\hbar}{p_F L}\right)^{9/2} \left(\frac{\sigma^2 p_F}{\hbar L}\right)^5 \, \infty \, \omega^3 \nu^{-8}(T) \,. \tag{22}$$

The width of the hysteresis loops of the induced magnetic moment as a function of the external magnetic field  $h_0$  increases linearly  $(\Delta h_0 \approx 4\mathcal{H})$  in the range where the hysteresis is well developed, i.e., when

$$\mathcal{H}_{cr} \ll \mathcal{H} \ll \mathcal{H}_{cr}'.$$

When  $\mathcal{H}$  approaches  $\mathcal{H}'_{cr}$ , the width of the loops decreases abruptly. It was precisely this sort of dynamics in the variation of the hysteresis loop width with increasing wave amplitude that was observed experimentally.<sup>15</sup>

If the quality of the surface is such that the quantity  $(\sigma^2 p_F / \hbar L)^{1/6} \sigma / L$  is slightly greater than  $\delta_0 / l$ , then hysteresis does not develop. In this case the induced magnetic moment loops exist in a small range of amplitude near  $\mathcal{H}_{max}$ . The frequency dependence of  $\mathcal{H}_{max}$ , near which hysteresis occurs ( $\mathcal{H}_{max} \propto \omega^{1/3}$ ) agrees with that observed in Ref. 15.

Let us note that expressions (20)-(22) make it possible to determine the surface characteristics from the experimentally observed values of the amplitudes  $\mathcal{H}_{cr}$ ,  $\mathcal{H}_{max}$ , and  $\mathcal{H}'_{cr}$ : we can consider these expression as three equations in the three unknowns  $\sigma$ , L, and v. The authors wish to thank É. A. Kaner, L. M. Fisher, and I. F. Voloshin for helpful discussions.

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