Theory of a quasiatomic gas

A. A. Likal'ter

Institute of High Temperatures, Academy of Sciences of the USSR

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The transition between an atomic gas and a metal near the critical point is described through a model of the intermediate state of the substance: a quasiatomic gas. The most characteristic electronic properties of this state—its electrical conductivity, dielectric polarization, screening, and plasma oscillations—are discussed.

1. INTRODUCTION

By compressing the vapor of a metal, e.g., cesium or mercury, at temperatures in the critical region (~2000 K) one can observe a transition of an atomic gas into a metallic state. The latter is signalled by an increase in the electrical conductivity to $\sim 10^2$ S/cm and a weak temperature dependence of this conductivity (a Mott transition). The nature of the different phases of metals near the critical point, at which the difference between phases disappears, has attracted interest for a long time.[1] According to recent data, the transition of a vapor to a metallic state is continuous. It is nevertheless generally assumed that a Mott transition coincides with the point at which the activation energy for the electrical conductivity, found from the temperature dependence, disappears. The density of mercury at this point is higher than the critical density, while that of cesium is slightly lower.[2,3] Mercury and cesium thus constitute examples of the two types of phase diagrams for metals which were discussed in Ref. 1 (reviewed in Refs. 4, 5).

In contrast with thermal ionization, which results from excitation of atoms, near a Mott transition valence electrons of ground-state atoms are delocalized. It is thus difficult to distinguish atoms and free electrons and ions as separate subsystems. The atoms are in quasistationary states in which the valence electrons can go off to infinity (quasiatoms). A quasiatomic state refers to a certain atom, and the propagation of electrons is a random walk among neighboring atoms. Percolation theory is of fundamental importance for this problem. The model of a quasiatomic gas thus leads to a percolation theory of a Mott transition. The concept of quasiatoms was introduced in our papers in order to describe the electronic properties of a metal vapor near a Mott transition.[6] In the present paper we summarize the physical ideas underlying the model, and we examine the most characteristic properties of a quasiatomic gas: its electrical conductivity, dielectric polarization, screening, and plasma oscillations. Only the first of these properties (and the most important) was studied in Ref. 6.

2. PERCOLATION CLUSTERS OF ATOMS

In atomic gas, unlike a metal, the potential wells in which the valence electrons move are separated by barriers. A point of importance for conversion to a metallic state is the ability of metal atoms to form percolation clusters in which valence electrons move among atoms above potential barriers. A necessary condition here is that the atoms close on each other to within a distance of $2e^2/I$ ($-e$ is the charge of an electron, and I the ionization potential of the atom) so there will be an overlap of the classically accessible regions for the motion of the valence electrons. Outside these regions, the fields of the ions are screened almost completely. For metals which have an *ns* valence shell with a large main quantum number one can use a semiclassical model for the screening of the atomic core by the valence electrons near the boundary of the classically accessible region. The potential produced by an atom outside this region is zero, while that inside the region (but outside the ion core) is $e/r - I/e$, where r is the distance from the nucleus. In the zone in which the classically accessible regions overlap, the potential energy of a valence electron of the atom has a saddle point, at which it reaches the value $-4e^2/R + I$, where $R < 2e^2/I$ is the distance between the overlapping atoms. The potential barrier between the overlapping atoms thus lies below the ground state of the atom.

If the relative value of the classically accessible volume of the atoms,

$$\zeta_0 = \frac{4\pi}{3}\left(\frac{e^2}{I}\right)^3 n_a \tag{1}$$

reaches a threshold $\zeta_t \approx 1/3$ (this is the percolation threshold in the problem of overlapping spheres[7]), an infinite percolation cluster forms. The percolation threshold is naturally identified with a Mott transition (in mercury, this threshold is $\zeta_t = 0.29$). The idea that the conversion to a metallic state is linked with a percolation among atomic spheres "filled" by the wave function has been expressed in several places (e.g., Ref. 8).

The clustering of atoms is important above the transition point and also in a certain neighborhood below it. According to the scaling theory,[9] the number of percolation clusters of s particles is described by the distribution

$$n_s \propto s^{-\tau}, \quad \tau = 2.15.$$

Here $s \ll s_\xi$ where s_ξ is a correlation parameter which diverges at the point of the transition. The power-law distribution reflects the fact that there are no characteristic values for the number of particles in a cluster at a scale much less than s_ξ. The integral for the number of atoms in clusters,

$$\int_1^s s n_s \, ds \propto \frac{1 - s^{2-\tau}}{\tau - 2},$$

does converge at large s, but very slowly. Near the transition point, most of the atoms thus become part of large percolation clusters. Above the transition point, a finite fraction of the atoms is in an infinite cluster.

The correlation parameter diverges at the transition point[9] in accordance with

$$s_\xi \propto |\zeta_0 - \zeta_t|^{-1/\sigma}, \quad \sigma = 0.45.$$

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We thus see that the clustering condition $s_\xi \gg 1$ holds below the transition point if $\zeta_t - \zeta_0 < 0.1$.

3. SCREENING OF THE INTERACTION

Above-barrier transitions of electrons in percolation clusters lead to a screening of the ions by the electrons of neighboring quasiatoms. There is a finite probability for a valence electron to be far (in comparison with e^2/I) from its ion. The screening charge is related to the potential which it creates by the Poisson equation,

$$\frac{1}{r^2}\frac{d}{dr}r^2\frac{d\varphi}{dr}=-4\pi\rho(r). \qquad (2)$$

The distribution of the screening charge is normalized by the condition

$$\int_0^{R_s} 4\pi\rho r^2\,dr=-e,$$

where R_s is the total-screening radius. Integrating (2), we find

$$\varphi(R_s)-\varphi(0)=-\frac{e}{R_s}-4\pi\int_0^{R_s}\rho r\,dr. \qquad (3)$$

If a valence electron is near its ion, there is no external screening charge. We will make use of that circumstance in choosing a normalization of the potential: $\varphi(0) = 0$. Adding to (3) the potential of the ion, we find the asymptotic expression for the screened potential as $r \to \infty$:

$$\frac{e}{r}+\varphi(r)\sim-4\pi\int_0^{R_s}\rho r\,dr.$$

In a strongly interacting system, the screening will be nonlinear, so the distribution of the screening charge cannot be determined explicitly. It is natural to suggest, however, that this charge will be about the same as in the atom. Assuming

$$4\pi r^2\rho(r)=-e\delta(r-e^2/I),$$

we find the asymptotic value of the screened potential, I/e; this value corresponds to $-I$ as the potential energy of the electron. The potential energy of the electron close to the ion is $-e^2/r$ in this case.

The sum of the screened potential of the electron's ion and of the potentials created by the atoms generates a muffin-tin potential-energy relief: a collection of Coulomb wells lying below a plateau of height $-I$. In this potential (which has no bound states) an electron moves far from its ion, nearly as a free electron would in the sense of the theory of metals.[8]

4. QUASIATOMIC STATES

As a result of the screening, there is a certain probability that the valence electrons will be in free states, while the atomic states are quasistationary. The interaction mixes these states. The internal-energy spectrum of an atom in mixed (quasiatomic) states corresponds to an asymptotically free motion of an electron, and according to the variational principle it lies above the ground level of the free atom. Treating the change in the state of an atom in a percolation cluster as a small perturbation, we can minimize the corresponding change in internal energy. The internal-energy spectrum of a quasiatom is then

$$E=-I+p^2/2m, \qquad (4)$$

where p is the momentum of the free motion, and m is the mass of the electron.

We represent the quasiatomic states by the density matrix

$$\rho(x,x')=a_{00}\Psi_0^*(x')\Psi_0(x)+a_{pp}\Phi_p^*(x')\Phi_p(x), \qquad (5)$$

where Ψ_0 is the wave function of an electron in the ground state of the atom, Φ_p is the wave function of the continuum, and a_{00} and a_{pp} are the diagonal elements of the energy representation of the density matrix (a_{00}, $a_{pp} > 0$, $a_{00} + a_{pp} = 1$). Equating the internal energy $E = a_{00}(-I) + a_{pp}(p^2/2m)$ to expression (4), and using the normalization of the matrix elements, we find

$$a_{pp}/a_{00}=p^2/2mI. \qquad (6)$$

It follows from (5) and (6) that weakly excited quasiatomic states with $p^2/2mI \ll 1$ differ little from the ground state (except in terms of asymptotic behavior).

For finite percolation clusters one can speak in terms of an intermediate asymptotic behavior of the density matrix and of a free motion of electrons in a bounded region. The spectrum of a quasiatom is continuous if the uncertainty in the energy, $\hbar/\tau$, where τ is the mean free time of an electron, is greater than the exchange splitting of the terms of the quasimolecule at an interatomic distance $\sim 2e^2/I$.

We introduce the average velocity of the free motion of the electrons, v_e, and the average time for a transition of a valence electron to another quasiatom, τ'. The typical radius of the quasiatomic states is then $v_e\tau'$, and the volume which corresponds to them is

$$\Omega=\frac{4\pi}{3}(v_e\tau')^3=\frac{1}{n_i}\left(\frac{\tau'}{\tau}\right)^3. \qquad (7)$$

Here $\tau = l/v_e$, $l = (4\pi n_i/3)^{-1/3}$ is the average radius of an ion cell, and n_i is the density of ions. Since the relation $\tau' \gg \tau$ holds for quasistationary states, the volume which corresponds to them, Ω, is much greater than the volume of an ion cell. Over the greater part of Ω an electron moves in a screened potential, i.e., nearly as a free electron. In the approximation of free motion, the state density in the internal-energy spectrum of the quasiatom is described by

$$\frac{dg}{dp}=g_0\frac{4\pi p^2\Omega}{(2\pi\hbar)^3}, \qquad (8)$$

where g_0 is the statistical weight of the ground level of the atom.

The nature of the electron energy distribution depends on the relation between the temperature and the Fermi energy $\varepsilon_F' = p_F'^2/2m$. The Fermi momentum in a quasiatomic gas is determined by the state density (8) and is given by

$$p_F'=\hbar\left(\frac{6\pi^2 n_i}{g_0}\right)^{1/3}\frac{\tau}{\tau'}. \qquad (9)$$

In the case of *ns* valence shells we would have $p'_F = p_F\tau/\tau'$, where $p_F = \hbar(3\pi^2 n_e)^{1/3}$ is the Fermi momentum of a homogeneous electron gas. Since the relation $\tau \ll \tau'$ holds, the Fermi energy in a quasiatomic gas is much lower than that in a homogeneous electron gas of the same density. The range of applicability of Boltzmann statistics is significantly extended.

5. DIFFUSION AND MOBILITY OF ELECTRONS

The center of the wave packet which corresponds to a valence electron undergoes a random walk between the overlapping atoms. The electron diffusion coefficient is

$$D_e \approx \frac{l^2}{3\tau'} = \frac{1}{3} l v_e \frac{\tau}{\tau'}. \tag{10}$$

The difference between (10) and the expression from the kinetic theory of gases with a minimum range l, according to Ioffe and Regel',[4] lies in the factor τ/τ', which is a measure of the partial localization of the electrons.

Correspondingly, for the mobility we have

$$\mu = \frac{e\tau}{m}\frac{\tau}{\tau'}. \tag{11}$$

Expressed in terms of the formulas from the kinetic theory of gases, the diffusion coefficient and the mobility obviously satisfy the Einstein relation.

Expression (11) shows that the electrons drift in an electric field $\mathscr{E}$ as free electrons would drift in an effective field $\mathscr{E}\tau/\tau'$. In other words, the effect of the external field on the propagating electrons is weakened by the localization factor. This point is of importance to the screening of the external field and the plasma oscillations (more on this below).

In a degenerate quasiatomic gas we have $\tau = l/v'_F$. Substituting this value of τ into (11), and expressing p'_F in terms of p_F, we find

$$\mu = el/p_F. \tag{12}$$

The factor τ/τ' is thus cancelled out in (12). The mobility given by (12) corresponds to the so-called minimum metallic electrical conductivity.[7] In the Boltzmann case we have $\tau = l/v_e$; the average velocity $v_e = (8T/\pi m)^{1/2}$ depends only on the temperature T. Under the condition $\tau \ll \tau'$, the mobility given by (11) is thus much lower than the kinetic-theory mobility.

We now find an explicit expression for the localization factor τ/τ'. For this purpose we consider how the mobility μ_p of electrons with a free momentum p depends on the excitation which results from the increase in the relative size of the classically accessible volume. For an electron which is undergoing a transition from one atom to another and which has an excitation energy $p^2/2m$, the relative size of the classically accessible volume is

$$\zeta_p = \frac{\zeta_0}{(1-p^2/2mI)^3}, \tag{13}$$

where ζ_0 is given by (1). We set

$$\mu_p = (e\tau/m) f(\zeta_p),$$

where the function $f(\zeta_p)$ is zero for $\zeta_p < \zeta_t$ and one for $\zeta_p > \zeta_f$, and ζ_f is a scale value of the relative size of the classically accessible volume in the case of free propagation (e.g., the degree of spherical close packing, 0.74). We then have

$$\tau/\tau' = \langle f(\zeta_p)\rangle, \tag{14}$$

where the angle brackets denote the average over a Boltzmann distribution. We formally expand $f(\zeta_p)$ in the small parameter $p^2/2mI$. Introducing a percolation level Δ_1 and a free-propagation level Δ_2 (putting the origin for the energy scale at $-I$), we write

$$f(\zeta_p) = \begin{cases} 0, & p^2/2m < \Delta_1 \\ (p^2/2m - \Delta_1)(\Delta_2 - \Delta_1)^{-1}, & \Delta_1 < p^2/2m < \Delta_2. \\ 1, & p^2/2m > \Delta_2 \end{cases} \tag{15}$$

The parameters Δ_1 and Δ_2 are expressed in terms of the typical values of the relative size of the accessible volume. From (13) we find

$$\Delta_1 = I[1-(\zeta_0/\zeta_t)^{1/3}]$$

and a corresponding expression for Δ_2, in which ζ_t is replaced by ζ_f. At the transition point, Δ_1 vanishes.

Using (15), and taking an average over a Boltzmann distribution in (14) with state density (8), we find[6] an exponential temperature dependence for $\zeta_0 < \zeta_t$:

$$\frac{\tau}{\tau'} \approx \frac{2}{\pi^{1/2}} \frac{\Delta_1}{\Delta_2 - \Delta_1} \left(\frac{T}{\Delta_1}\right)^{1/2} \exp(-\Delta_1/T), \quad \frac{\Delta_1}{T} \gg 1. \tag{16}$$

For $\zeta_0 > \zeta_t$ we find a linear dependence:

$$\frac{\tau}{\tau'} \approx \frac{3T/2 - \Delta_1}{\Delta_2 - \Delta_1}, \quad \frac{\Delta_2}{T} \gg 1. \tag{17}$$

The change in the nature of the temperature dependence in (16) and (17) when the mobility threshold disappears coincides with the Mott transition. The volume of quasiatomic states, (7), is found with the help of (17) [below the transition point, the value of τ/τ' at this point itself, which corresponds to the local density in the region spanned by the percolation cluster, appears in (7)].

Expressions (11), (16), and (17) give the solution of the electron-mobility problem. Note that the mobility is not the same as the mobility due exclusively to the finite probability for a free motion of an electron above the transition point:

$$\mu \gg \frac{e\tau}{m}\langle a_{pp}\rangle, \quad \langle a_{pp}\rangle \simeq \frac{3T}{2I}.$$

The electrical conductivity is

$$\sigma = \frac{e^2 n_e \tau}{m}\frac{\tau}{\tau'},$$

where n_e is the density of valence electrons. Figure 1 shows results calculated for the electrical conductivity of cesium and mercury.

The rf conductivity in the Drude approximation is

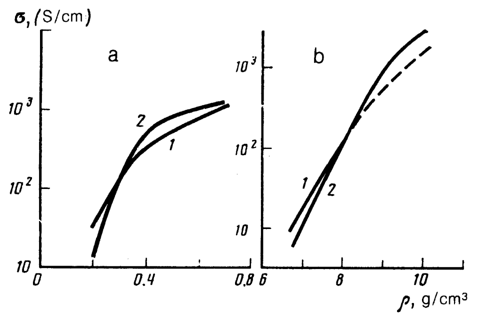

FIG. 1. Electrical conductivity of (a) cesium at $T = 2113$ K and (b) mercury at $T = 1800$ K. 1—Experimental[2,3]; dashed line—extrapolation over temperature; 2—calculated with an electron localization factor τ/τ'.

$$\sigma(\omega)=\frac{e^2n_e\tau}{m}\frac{\tau}{\tau'}\frac{1}{1-i\omega\tau}.$$

At optical frequencies, the localization factor τ/τ' depends on the frequency as a result of the excitation of a quasiatom when the virtual absorption of a photon $\hbar/\omega$ occurs. For photon energies which are not too large, $\hbar\omega \ll \Delta_2$, this effect is described by the replacement $\Delta_1 \to \Delta_1 - \hbar\omega$, $\Delta_2 \to \Delta_2 - \hbar\omega$. Using this replacement, we find an exponential frequency dependence for the dynamic localization factor below the point of the Mott transition: $\tau/\tau' \propto \exp(\hbar\omega/T)$. Figure 2 shows results calculated for the optical conductivity of mercury.

6. DIELECTRIC POLARIZATION

The polarization of metal atoms which have ns or ns^2 valence shells with large n reduces to a shift of the electron charge in the electric field along with the classically accessible region of motion. The boundary of the classically accessible region is described by the equation

$$-e^2/r-e\mathcal{E}r\cos\theta=-I$$

(the axis of the spherical coordinate system runs opposite the external field $\mathcal{E}$). A weak field $\mathcal{E} \ll I^2/e^3$ causes a shift of the points of the boundary sphere in the radial direction by an amount $\delta r = (e^5/I^3)\mathcal{E}\cos\theta$, which corresponds to a shift of the entire sphere along the z axis by an amount $(e^5/I^3)\mathcal{E}$. Multiplying this shift (at a unit field) by e and by the number of valence electrons, z, we find the polarizability:

$$\alpha_a=z(e^2/I)^3. \qquad (18)$$

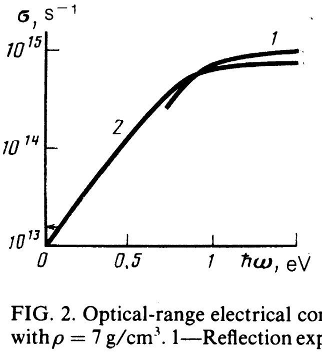

FIG. 2. Optical-range electrical conductivity of mercury at $T = 1800$ K with $\rho = 7$ g/cm³. 1—Reflection experiment[10] (the arrow shows the static conductivity); 2—calculated with a dynamic localization factor.

This expression is an approximate description of the polarizability of both free atoms and quasiatoms in percolation clusters if the classically accessible regions have been determined well.

In addition to the quasiatoms themselves in percolation clusters, the bonds between overlapping quasiatoms have become polarized; i.e., a partial transition of an electron between the quasiatoms may be induced by the electric field. In a weak field, the polarization of individual bonds can be treated independently.

The energy of a quasimolecule goes through a maximum as a function of the degree of polarization δ at $\delta = 0$. The expansion of the excitation energy near the minimum begins with the term $\propto \delta^2$. We find the coefficient from the condition that the value $\delta = 1$ corresponds to the excitation energy of the ion term, $I - \varepsilon - e^2/R \approx I/2$ (here $\varepsilon \ll I$ is the electron affinity of the atom, and $R \approx 2e^2/I$ is the interatomic distance).

The change in the energy due to the polarization of the quasimolecule in the electric field is

$$\tfrac{1}{2}I\delta^2-2\delta e\mathcal{E}(e^2/I)\cos\theta$$

where θ is the angle between the direction of the electron transition (the axis of the quasimolecule) and the polar axis, which is directed opposite the external electric field. Minimizing this expression, we find

$$\delta=2(e^3/I^2)\mathcal{E}\cos\theta.$$

Evaluating the projection of the dipole moment onto the field, and taking an average over angles with the help of the relation $\langle\cos^2\theta\rangle = 1/3$, we find the polarizability of the bond:

$$\alpha_b=\tfrac{4}{3}(e^2/I)^3. \qquad (19)$$

The only difference between the polarizability of the bonds, (19), and that of the atoms, (18), is in the numerical coefficient. Combining the two, we introduce an effective polarizability of the quasiatom:

$$\alpha=(z+\tfrac{4}{3}c)(e^2/I)^3, \qquad (20)$$

where $c \sim 1$ is the average number of bonds per atom.

In calculating the electric susceptibility due to the polarization of the quasiatoms (and bonds), we need to consider the local-field effect. This effect can be summarized by saying that the external field for a structural element of a medium is not the average field itself but the average field minus the depolarizing field inside this element. In the approximation of a continuous medium, the latter field corresponds to the depolarizing field of a dielectric sphere, $-4\pi\chi\mathcal{E}/3$, where χ is the susceptibility of the medium. The field which determines the polarization of the quasiatom is $(1 + 4\pi\chi/3)\mathcal{E}$, so we arrive at the Clausius-Mosotti equation for the dielectric constant. Using (20) and definition (1), we find

$$\varepsilon-1=\frac{3(z+\tfrac{4}{3}c)\zeta_0}{1-(z+\tfrac{4}{3}c)\zeta_0}. \qquad (21)$$

The divergence in the Clausius-Mosotti equation corresponds to a spontaneous polarization, which is actually impossible in a homogeneous and isotropic medium. This divergence stems from the mean-field approximation, which overestimates the polarization of the atoms in one direction. Equation (21) is thus applicable not too close to a pole.

Well above the transition point at $\tau \sim \tau'$, the polarization of the quasiatoms and the corresponding polarization evidently vanish.

7. SCREENING AND PLASMA OSCILLATIONS

In addition to the dielectric polarization at the atomic scale, the electrons in a quasiatomic gas are polarized at distances much greater than the interatomic distances, screening the external field. The electron density in a self-consistent field with a potential φ is described by a Boltzmann distribution $n \propto \exp(e\varphi\tau/T\tau')$, which contains the effective potential $\varphi\tau/\tau'$. The linearized Poisson equation $\Delta\varphi = \varkappa^2\varphi$ describes an exponential screening of a potential $\varphi \propto e^{-\varkappa r}$. Here the reciprocal screening radius (the renormalized electron Debye length) is

$$\varkappa = \left(\frac{4\pi e^2 n_e}{\varepsilon T}\frac{\tau}{\tau'}\right)^{1/2}, \tag{22}$$

where ε is the dielectric constant which is associated with the polarization at the atomic scale.

The presence of a factor τ/τ' in (22) means that the screening radius depends on the degree of localization and is determined by electrons with energies above the mobility threshold. On the dielectric side of the transition, the radius of the screening of the external field depends exponentially on the density and decreases to the interatomic distance near the transition point.

If, for some reason, the electrical conductivity is negligible at the macroscopic scale, percolation clusters will be conducting inclusions which screen the external field from their interior. The correlation length ξ (the maximum size of a cluster), which determines the scale, diverges at the point of the transition[9] in accordance with

$$\xi \propto |\zeta_0 - \zeta_t|^{-\nu}, \quad \nu = 0.84.$$

When an infinite cluster forms, the external field is screened completely from the medium. In this case the medium constitutes a dielectric whose dielectric constant diverges at the point of the Mott transition. This effect, which is observed in extrinsic semiconductors at low temperatures, is known as the polarization catastrophe[11] (although it is not related to spontaneous polarization, as in the Clausius-Mosotti case). At high temperatures the effect is masked since the external field is again screened from the medium on the dielectric side of the transition.

As usual, plasma oscillations with a wavelength much greater than interatomic distances are associated with the screening. The equation for the drift motion of an electron in the electric field is

$$\frac{dv}{dt} = \frac{-e\mathscr{E}}{m}\frac{\tau}{\tau'} - \frac{v}{\tau}. \tag{23}$$

Combining (23) with the continuity equation $\partial q/\partial t - en_e \nabla v = 0$ ($q = -e\delta n_e$ is the charge density) and the Poisson equation, $\nabla\mathscr{E} = 4\pi q/\varepsilon$, we find an equation for the plasma oscillations:

$$\frac{\partial^2 q}{\partial t^2} + \omega_p^2 q + \frac{1}{\tau}\frac{\partial q}{\partial t} = 0, \tag{24}$$

where

$$\omega_p = \left(\frac{4\pi e^2 n_e}{\varepsilon m}\frac{\tau}{\tau'}\right)^{1/2} \tag{25}$$

is the renormalized plasma frequency. The damping rate is $1/2\tau$.

Formally, the usual relation $\omega_p \sim \varkappa v_e$ holds between the screening radius (22) and the plasma frequency (25). The situation changes when the finite energy of a plasmon, $\hbar\omega_p$, is taken into account. As for the optical-range electrical conductivity, the factor τ/τ' itself depends on ω_p (because of the virtual absorption of plasmons by quasiatoms), so (25) is actually an equation for ω_p. Below the point of the Mott transition, Eq. (25) can have one or two stable roots. At the lower frequency, which corresponds approximately to the static value τ/τ', oscillations are not possible because of the strong damping. In reality, plasma oscillations with a frequency corresponding to $\tau/\tau' \sim 1$ can be excited [in this case, the dielectric constant in (25) is $\varepsilon \sim 1$].

8. CONCLUSION

From the general point of view, a metal vapor near the critical point is a dense, strongly interacting plasma which converts into a metal upon compression, while an expansion converts it into an atomic or partially ionized gas. The ground state of the atoms in the intermediate region is a quasistationary state because of screening, so there is a finite probability for the delocalization of valence electrons. A fully microscopic description of a dense plasma of this sort is not possible because of the strong interaction. A phenomenological description is possible because a main interaction—that of the electron with its ion—can be distinguished. On this basis, the changes in the electronic properties at the transition are described with the help of percolation theory.

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