Nonlinear conductivity of a 3D lattice of GaAs clusters in opal

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(Submitted 23 March 1995)
Pis'ma Zh. Éksp. Teor. Fiz. **61**, No. 9, 738–742 (10 May 1995)

A 3D lattice of GaAs clusters (~500 Å) in an opal matrix has been synthesized. The current–voltage characteristic of the system has regularly spaced slope changes which are the boundaries between essentially straight regions. These features of the current–voltage characteristic can be interpreted as resulting from the successive opening of various inelastic channels, accompanied by the emission of optical phonons whose energy increases in proportion to the square of the number of the branch. The structure can be thought of as a 3D lattice of nanostructures: tunnel junctions formed by virtue of the appearance of barriers between clusters (a 3D nonlinear medium). With a further increase in the voltage, there is a transition to a regime of a current limited by space charge concentrated near discontinuities in regions of the SiO_2 matrix. © *1995 American Institute of Physics.*

The synthesis of regular 3D lattices of clusters, ranging in diameter from 10 to 1000 Å, makes possible a transition from ordinary crystals, with lattice constants of about 3 Å (a case which has been studied thoroughly), to crystals, scaled up by a factor of 10–20 and to regular 3D arrays of nanostructures. Cluster crystals (based on clusters in the voids of zeolites, for example) have some extremely prominent nonlinear properties even in weak electric fields.[1] These systems constitute 3D versions of superlattices. An even greater increase in the scaling factor (to 100–1000) makes possible a switch from systems with a common electronic energy structure (i.e., from crystals) to nanocomposites with the intrinsic electronic and phonon systems of the components which are interacting with each other and also to 3D lattices of nanostructures or media (e.g., to systems of Josephson type[2] or to lattices of thermionic converters[3] based on opals). There is a transition region between these two types of systems.

We have synthesized GaAs clusters in the voids of artificial opals with a perfect structure. Figure 1 shows a fragment of the structure of the lattice of voids in an opal. The dashed lines show spheres of the SiO_2 matrix of the opal, of radius R. The other dimensions here are

$$r_1 = R(\sqrt{2}-1) = 0.41R, \quad r_2 = R\left(\sqrt{\frac{3}{2}}-1\right) = 0.225R,$$

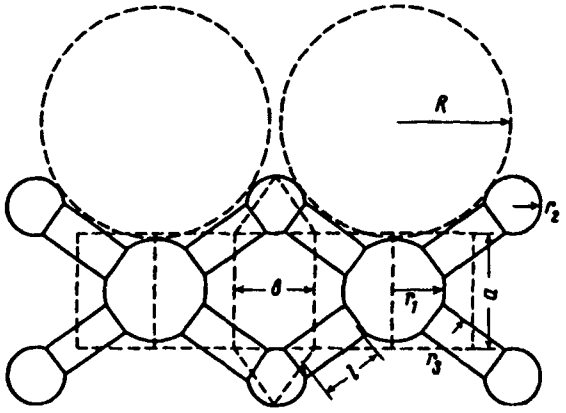

FIG. 1. Structural elements of a lattice of voids in opal. These elements lie in a common plane.

$$r_3 = R\left(\frac{2}{\sqrt{3}} - 1\right) = 0.155R, \quad l = R(2-\sqrt{2}) = 0.59R.$$

Here a and b are the sizes of the cubes and tetrahedra. The system of voids in the opal can also be represented as a lattice of these cubes and tetrahedra.

The voids of the opal are 60% filled with gallium arsenide. In an opal, GaAs has an n-type conductivity. From measurements of the conductivity $\sigma = 0.02$ S/cm we find $\Delta E \approx 0.4$ eV, which corresponds to data on bulk GaAs. Silicon can serve as a dopant which can be introduced from a highly textured surface of a contact with the matrix. We see in Fig. 1 that the degree of doping (n_1) of the thin channels can evidently be higher than that (n_2) of the clusters, because of the greater role played by the surface. In the case of a lattice of HgSe clusters, which we have also synthesized in an opal matrix, we found $n_1 \approx 4n_2$ (this is a lattice of isotype n-n junctions connected in opposite directions).

Current–voltage characteristics were measured with the help of square pulses with a duration $\tau = 0.2$ μs, in a region in which the shape of the current–voltage characteristics was unaffected by either the value of τ or the duty factor.

Figure 2 shows the relative conductivity versus the reciprocal of the electric field. There is a series of linear regions, whose boundaries are at fields of 1140, 3450, 7060, 13 000 V/cm. As the field is raised, the current–voltage characteristic is described by $I \propto E^{3/2}$. Ohm's law holds up to 1140 V/cm. The inset in Fig. 2 shows $\sqrt{E_k}$ as a function of k, the index of the region. The condition $E \propto k^2$ holds well. The curves of $I(E)$ which we found for corresponding cluster lattices of Te and HgSe have fewer changes in slope and an earlier transition to an $I \propto E^{3/2}$ behavior.

This behavior indicates that there are threshold features in the current transport through this synthetic periodic medium: As the applied voltage is increased, new transport channels open up in succession. The sharpness of the features indicates that the process is of a quantum-mechanical nature; in the case of classical statistics, there could be only a smooth crossover. There may be disruptions of direct electronic contact be-

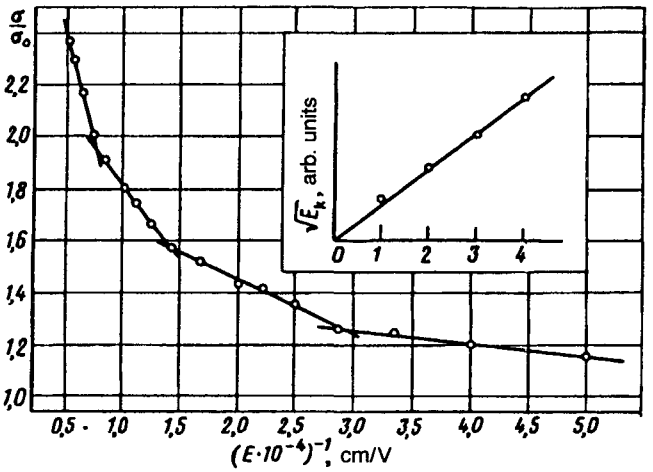

FIG. 2. Relative conductivity of a lattice of GaAs clusters in opal versus the reciprocal of the electric field E. The inset shows $\sqrt{E_k}$ versus k, which is the index of the slope change.

tween different GaAs clusters in these structures. The most probable locations of such disruptions are the narrow channels connecting large clusters. So far, we do not have any convincing data regarding the number or distribution of these discontinuities. It seems extremely likely that they would be arranged as a regular lattice involving two discontinuities per unit cell of the opal. In the case of a large tunneling resistance we could assume that the voltage V applied to the sample drops primarily across the barriers. If so, the voltage across one barrier, U, can be calculated quite easily under the assumption that the voltage is distributed uniformly among the barriers: $U=V/m$, where m is the number of barriers along current lines, which is in turn equal to $N\nu$, where N is the number of opal cells, and ν is the number of barriers in a cell along the current lines. Under these assumptions, detailed information on the nature and structure of the barriers is important for the magnitude of the current, but not for determining the positions of the features on the current–voltage characteristic along the axis of the applied voltage. Note that this structure is a new entity—a 3D-periodic lattice of tunnel junctions of mesoscopic scale—and this study is merely a first step in research on this entity. Our task is therefore one of analyzing the most prominent features of the observed phenomena. One of these phenomena is the presence of slope changes on the current–voltage characteristic, which occur in a definite sequence: The distance between slope changes increases with the index of the slope change, k (Fig. 2).

The current–voltage characteristic consists of three characteristic parts: an ohmic part, a piecewise-linear part (despite the linearity of the individual pieces of the current–voltage characteristic, it cannot be thought of as ohmic), and a power-law part at high voltages ($I \propto V^{\alpha}$, where $\alpha=3/2$). The following seems to be the most probable explanation of the observed picture: The ohmic region at low voltages is determined by a drift of carriers injected by the semiconductor clusters into the insulating regions. The resistance is dominated by the tunnel junctions, which are operating in a regime of elastic tunneling.

This contribution to the resistance can be estimated from the Landauer formula:[4]

$$R=\frac{h}{e^2}\frac{1-T}{T},$$

where T is the transmission coefficient of the barrier for an electron wave, h is Planck's constant, and e is the charge of an electron. As the applied voltage is raised, inelastic scattering channels come into play, and optical phonons in the matrix are emitted. The inelastic increment in the current which arises in the process is proportional to a statistical factor of the type[5]

$$I\propto\int_{-\infty}^{+\infty}f(E)[1-f(E+eV-h\omega_0)]dE.$$

At $T\gg h\omega_0$ we have

$$I_{\text{inel}}\propto\begin{cases}0, & eV<h\omega_0,\\ (eV-h\omega_0)\dfrac{\exp[(eV-h\omega_0)/kT]}{\exp[(eV-h\omega_0)/kT]-1} & eV>h\omega_0.\end{cases}$$

Raising the temperature smooths out the slope changes somewhat. The first derivative is a sequence of steps, while the second is a sequence of peaks. Peaks with approximately the same sequence ($\omega_k\propto k^2$) are observed[6] in the IR spectra of quartz beginning at $k=2$. These spectra reflect the density of optical vibrations of the crystal. Although we regard the interpretation offered above as the most likely one, we do not rule out other possibilities. For example, there might be a resonant tunneling between neighboring wells,[7] or there might be an electrophonon resonance.[8,9] Each of these two mechanisms would lead to peaks in the current itself, not in its second derivative. However, by choosing some specific tunneling probabilities, etc., we might achieve a theoretical current–voltage characteristic approaching that observed experimentally. Yet another possibility for tunneling between clusters would be a Coulomb blockade interrupted by inelastic processes. The question must be examined in detail, but this explanation does conform to the context of the original picture, with the Coulomb interaction being taken into account.

Some other important questions are just where these discontinuities are and just what the tunneling paths are.

There are numerous possibilities which may be realized, depending on the synthesis conditions. A first possibility, corresponding to a dense filling of the voids, including the channels, suggests that there are thin discontinuities in the material filling the channels, which allow electrons to tunnel through the surrounding insulating medium. A second possibility, corresponding to a less dense filling, is based on the assumption that there are some relatively long unfilled channels separating clusters. In this case one could speak in terms of a doping of the SiO_2 layers adjacent to the channels, by virtue of an adsorption of As atoms on the channel walls. Barriers would arise at the boundaries with clusters. There is the further possibility of an injection of carriers from clusters, which would allow electrons to move along the surface of a channel with a low resistance (in addition to the contact barriers between the clusters and the channels).

This system may be thought of as a 3D lattice of nanostructures of the Schottky-diode type, with dimensions on the order of 1000 Å , connected in opposite directions. Explanations of the features on the current–voltage characteristics which invoke an inelastic emission of optical phonons raise the hope that it would be possible to observe a change in the noise spectrum by virtue of the emission of optical phonons during tunneling. They also raise the hope of observing a possible modification of the current–voltage characteristic by applying an electromagnetic field of the appropriate frequency.

In a lattice of Te or HgSe clusters with a high carrier concentration, the transition to an $I=E^{3/2}$ law is observed earlier than in the case of GaAs. This result can be explained on the basis of the better injection capability of these clusters.

An $I=E^{3/2}$ law and a regime of a space-charge-limited current do not imply a tunneling through vacuum, but they can be explained in terms of a spherical geometry of the clusters with a spreading of the current in the matrix. Another reason might be the absence of a drift in the insulator because of the small dimensions of the barriers (a ballistic regime).

This study was supported, in part, by the International Science Foundation (Grant R1X000) and the Russian program Physics of Solid State Nanostructures (Grant 1-006).

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Translated by D. Parsons