Wigner crystallization and a resonance exchange mechanism for electrons localized in an amorphous insulator with a high trap density

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The possibility of Wigner crystallization of electrons in an amorphous insulator with a high trap density is discussed. A new exhange interaction mechanism is proposed, based on resonance tunneling of electrons between unfilled localized electronic states. © *1996 American Institute of Physics.* [S0021-3640(96)00719-0]

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A system of interacting electrons can become ordered into a regular Wigner lattice if the potential energy exceeds the kinetic energy.[1] The possibility of Wigner crystallization of electrons was analyzed in Ref. 2 for crystallization above the surface of liquid helium and in Ref. 3 for crystallization in the inversion channel of a semiconductor. In the present Letter a fundamentally different situation is examined—crystallization of electrons in deep traps in an insulator. In the case when the trap density N is high, so that the average distance $N^{-1/3}$ between traps is less than the effective trapping radius $(\sigma/\pi)^{1/2}$ (σ is the trapping cross section), electrons are trapped only in a fraction n_t of the traps, forming a quasiperiodic lattice. The period of this lattice is of the order of the characteristic Coulomb repulsion radius. If, moreover, the traps possess the same energy E, a phase transition—antiferromagnetic ordering on account of a resonance exchange interaction—can occur.

1. WIGNER CRYSTALLIZATION

We shall study a wide-gap insulator with no intrinsic or extrinsic mobile charge carriers. Deep, neutral, unfilled traps with high density N are present in the insulator. Consider a chain of traps separated by an average distance $a=N^{-1/3}$ (Fig. 1a). The localization energy E of the traps can be different. The distance between the traps can also be different. We shall study the trapping of electrons injected from the contact into the insulator. When an electron is trapped in trap A, a repulsive potential $e^2/4\varepsilon\varepsilon_0 R$, where ε is the dielectric constant,[a)] acts on a second (test) electron located at a distance R. A second electron with kinetic energy $E_{\rm ph}$ can approach the charged trap A to a distance $2R_q$ given by the relation

$$\frac{e^2}{4\pi\varepsilon\varepsilon_0 R_q}=E_{\rm ph}. \tag{1}$$

Hence we obtain the density n_t of filled traps, $n_t=(2R_q)^{-3}$:

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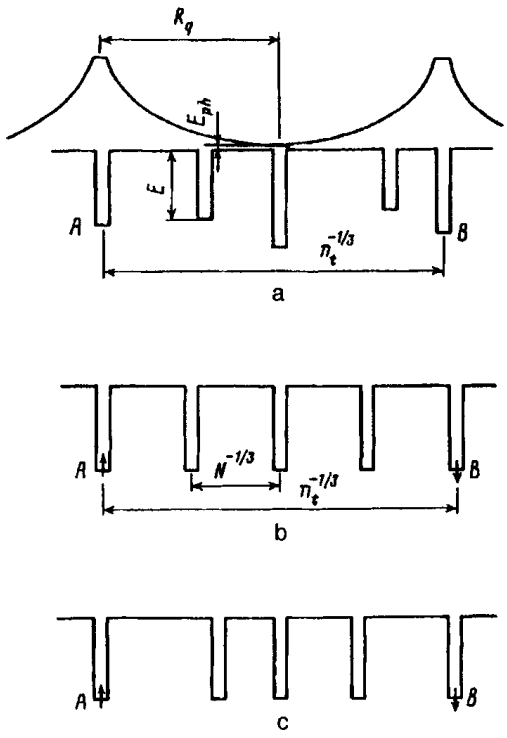

FIG. 1. Potential energy diagram illustrating the formation of a quasiperiodic electron lattice on account of Coulomb repulsion (a) and antiferromagnetic ordering in a periodic lattice on account of the resonance exchange interaction (b). Nonperiodic lattice (c).

$$n_t = \frac{(4\pi\varepsilon\varepsilon_0 E_{\rm ph})^3}{e^6}. \tag{2}$$

Figure 2 illustrates the formation of a two-dimensional square lattice with period $2R_q$. In our model such a lattice is formed by hard spheres with radius R_q. A gap of magnitude $N^{-1/3}$ can exist between the spheres. This quantity gives the characteristic scale of the variance in the lattice period.

Melting of a Wigner crystal occurs when $\Gamma = r_0/a_B^* \gg 1$. Here $a_B^* = 4\pi\varepsilon_0\varepsilon\hbar^2/m^*e^2$ is the effective Bohr radius.[2] For our model crystal, a phase transition will occur when the Coulomb interaction energy is greater than the binding energy E of an electron in a trap: $e^2 n_t^{1/3}/4\pi\varepsilon\varepsilon_0 \gg E$. Another obvious criterion is $E \gg kT$. This means that the ionization probability of the traps is low.

A universal parameter of the model is the characteristic Coulomb repulsion radius R_q. In our model R_q is greater than the distance between the traps: $R_q > N^{-1/3}$. The trapping cross section σ is determined not by the individual properties of a trap or the mechanism of energy loss, but rather by the square of R_q:

$$\sigma = \pi R_q^2 = \frac{\pi e^4}{(4\pi\varepsilon\varepsilon_0 E_{\rm ph})^2}. \tag{3}$$

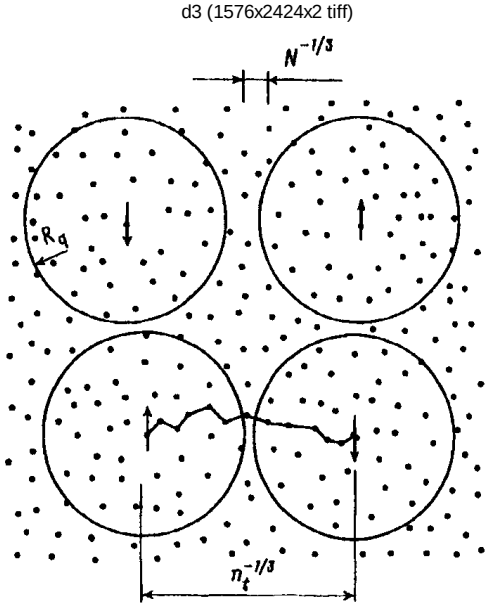

FIG. 2. Two-dimensional scheme of the formation of a quasiperiodic lattice on account of the Coulomb repulsion of electrons localized in traps and antiferromagnetic pairing of electrons on account of a resonance exchange interaction. A tunneling exchange trajectory is indicated for the bottom pair of electrons.

Therefore in our model the density of filled traps and the trapping cross section are related by the relation

$$n_t^{-1/3}/2=(\sigma/\pi)^{1/2}=R_q. \tag{4}$$

The quantity E_q in our model is the mean free path with respect to trapping: $R_q \simeq (n_t\sigma)^{-1}$. The trapping probability w for an electron in a layer of thickness R_q is $w=n_t^{2/3}\sigma=\pi$. Our model insulator can localize all electrons injected into it until the traps are ionized in the intrinsic space charge field.

In the present model the characteristic distance R_q between localized electrons is determined by the dielectric constant and the phonon energy. In our quasiperiodic lattice the period is determined, just as in a periodic Wigner lattice, by the Coulomb repulsion radius. A characteristic feature of our lattice is that the electrons are localized in deep states. Since traps are distributed randomly in the insulator, our lattice is quasiperiodic. We note that in our simple model one-, two-, and three-dimensional Wigner crystals can form. The criterion for crystallization in our model is relation (4).

In the case when all traps in the insulator have the same energy E and are located at the same distance a, another physical phenomenon occurs — antiferromagnetic pairing of localized spins.

2. ANTIFERROMAGNETIC ORDERING OF LOCALIZED ELECTRONS AS A RESULT OF A RESONANCE EXCHANGE INTERACTION

Let us examine the magnetic properties of electrons in a periodic superlattice formed by traps (Figs. 1b). The exchange integral J for electrons 1 and 2 at the sites A and B has the form[4]

$$J=\int\int \psi_A(1)\psi_B(2)\frac{e^2}{|\mathbf{r}_1-\mathbf{r}_2|}\psi_B(1)\psi_A(2)d\mathbf{r}_1 d\mathbf{r}_2, \tag{5}$$

where $\mathbf{r}_1$ and $\mathbf{r}_2$ are measured from the sites A and B, respectively. To calculate the exchange integral, the eigenfunctions for an electron in a lattice must be found. To illustrate the proposed exchange interaction mechanism, we shall not solve the exact problem of determining the stationary spin density in a superlattice. We shall confine ourselves to the WKB approximation. The probability P of finding an electron 1 of the well A at a distance R is

$$P=\exp\left(-\frac{2}{\hbar}R\sqrt{2m^*E}\right), \tag{6}$$

where m^* is the effective mass of the electron. For our model (Fig. 1b), E corresponds to the energy of the ground state in the well. The direct exchange interaction energy of the electrons at the sites A and B equals $q^2Pn_t^{1/3}/4\pi\varepsilon\varepsilon_0$.

Antiferromagnetic ordering will occur if the exchange interaction energy is greater than the Zeeman splitting of the level of a localized electron:

$$(e^2n_t^{1/3}/4\pi\varepsilon\varepsilon_0)\exp(-2n_t^{-1/3}/\hbar\sqrt{2m^*E})>g\mu H, \tag{7}$$

where μ is the Bohr magneton and H is the intensity of the magnetic field. The typical Zeeman splitting for electrons localized in a solid is $g\mu H\simeq 6\cdot 10^{-5}$ eV. For $n_t=10^{18}$ cm^{-3}, $\varepsilon=10$, and $E=1$ eV, we obtain the tunneling probability $P=10^{-27}$ and exchange interaction energy 10^{-29} eV (for $m^*=0.4m_e$).

Therefore the direct exchange interaction energy of electrons in traps is too low to cause a spin flip. The presence of a superlattice in our model results in delocalization of the spin density. We shall estimate the exchange interaction between an electron at site A and in a neighboring unfilled well. Substituting into Eq. (6) $R=N^{-1/3}=10^{-7}$ cm we obtain for the tunneling probability $p\simeq 10^{-3}$. The exchange interaction energy is $\simeq 10^{-4}$ eV. This is greater than the magnetic splitting. The estimate presented shows that in the presence of unfilled traps with high density the exchange interaction between electrons localized in traps can result in magnetic ordering of the spin subsystem.

In the present model there arises the question of whether or not total delocalization of electrons at the sites A and B occurs when a superlattice is formed. Analysis shows that the Coulomb interaction energy of electrons localized at the sites A and B equals the corresponding energy of the delocalized electrons.

We have studied a periodic superlattice. An exchange interaction can also occur in the case when the traps are distributed nonperiodically in space but have the same energy E (Fig. 1c). In this case the probability of an electron tunneling from the well A into the well B is not the product of the tunneling probabilities through the separate barriers. In

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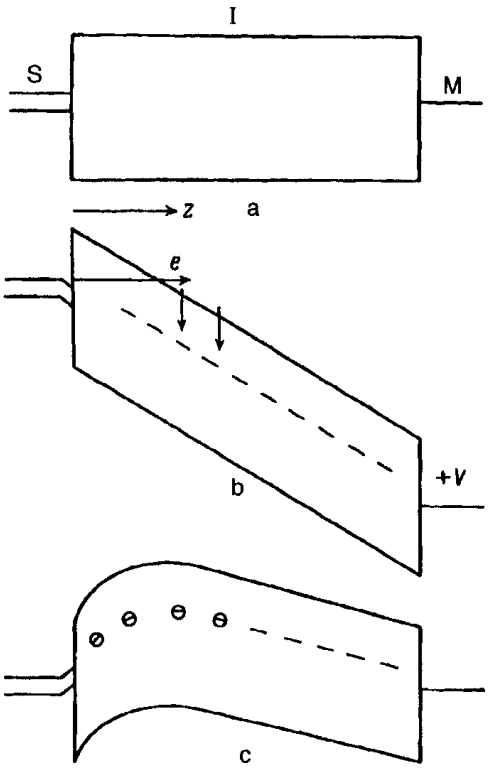

FIG. 3. Energy diagram of a MIS structure: a—Flat-band regime, b—injection of electrons from the semiconductor and trapping in traps in the insulator, c—shorted state.

the case when the barrier transmittances are substantially different the total tunneling probability is determined by tunneling between the most widely separated pair of traps (they correspond to the widest barrier). We note that for an exchange interaction between a pair of traps, an electron can choose a chain of traps with the minimum barrier width (Fig. 2). In this case a percolation problem arises. As follows from Fig. 2, a more realistic situation is binary pairing of spins in neighboring filled traps.

Amorphous insulators, employed in MIS (metal–insulator–semiconductor) structures,[6] can serve as objects for observing Wigner crystallization and antiferromagnetic ordering of electrons. The density of equilibrium intrinsic and extrinsic carriers in wide-gap amorphous insulators is negligibly low. For this reason, in contrast to doped semiconductors, a charge cannot be screened by free mobile charge carriers.[7] Electrons and holes localized in traps in the insulator in MIS structures are screened by mobile carriers in the semiconductor and metal.

The real situation for observing the above-indicated effects is more complicated. Figure 3 shows the energy diagram of a MIS structure in the flat-band regime (a), with the application of a field stimulating carrier injection from the semiconductor into the insulator (b), and in the shorted state (c). Carrier injection into the insulator can occur by tunneling, by photoemission, and as a result of heating in the depletion layer of the

semiconductor in the MIS structure.[7] Charge accumulation in traps results in the formation of space charge $en_t(Z)$ and the appearance of an electric field $e\int_0^z n_t(Z)dZ/\varepsilon\varepsilon_0$ in a direction normal to the plane of the MIS structure.

Wigner crystallization in the insulator in a MIS structure can correspond to the three-dimensional case. Antiferromagnetic ordering of the spins can occur only in a plane parallel to the plane of the semiconductor. In a direction normal to the surface, a trap is shifted in energy on account of the electric field by the amount $\Delta\psi = en_t^{-1/3}\int_0^z n_+(Z)dZ/\varepsilon\varepsilon_0$. For the typical intrinsic space-charge field in an insulator $e\int_0^z n_t(Z)dZ/\varepsilon\varepsilon_0 \simeq 10^6$ V/cm and $n_t = 10^{18}$ cm^{-3}, the displacement of neighboring traps in the z direction is $\Delta\phi = 1.2$ eV. This displacement prevents tunneling exchange in a direction normal to the surface. In a direction parallel to the plane of the semiconductor, there is no electric field because the localized charges are screened by the semiconductor and the metal.

The absence of ESR in a system with localized electrons can serve as a criterion of a resonance exchange interaction.

Several exchange interaction mechanisms are now known. The direct Heisenberg exchange mechanism explains ferromagnetic properties. The Anderson superexchange mechanism between d electrons via oxygen p states explains the ferromagnetic properties of oxides. The indirect Ruderman–Kittel exchange interaction describes the interaction between the localized spins of lattice ions via the conduction electrons of a metal.[4]

The resonance exchange mechanism proposed in the present Letter can explain antiferromagnetic ordering and absence of ESR in materials with a high density of localized states.

The analysis presented above was made for the case of electrons localized in an insulator. Experiments indicate that amorphous insulators can localize both electrons and holes.[8] All results obtained in the present work are also applicable to holes localized in an insulator.

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[a] In a polar insulator the polaron dielectric constant $\varepsilon_p^{-1} = \varepsilon_\infty^{-1} - \varepsilon^{-1}$ should be used for ε.

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