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Voltage-Induced Tunneling Conduction in Granular Metals at Low Temperatures

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We have observed a new conduction mechanism in granular metals at low temperatures and high electric fields $\mathcal{E}$ characterized by a field-dependent conductivity $\sigma_0 \exp(-\mathcal{E}_0/\mathcal{E})$. A theory based on a simple model of field-induced electron-hole generation in the bulk of the granular metal predicts the observed field dependence and gives expressions for σ_0 and $\mathcal{E}_0$ which are functions of the granular-metal parameters: metal grain size, and thickness and height of the tunneling barriers separating the metal grains. Agreement between theory and experiment is satisfactory.

We have observed a new electronic conduction process at low temperatures in granular metals[1] which consist of fine metallic particles separated by thin insulating tunneling barriers. Unusual transport effects result in these materials from the fact that the electrostatic energy E_c required to transfer an electron between two neutral grains can be appreciably larger than the thermal energy kT. Transport effects in granular metals at high temperatures and low electric fields have been studied by several workers.[2-4] The conduction model proposed by Neugebauer and Webb[2] in this regime is transport of electrons and holes due to tunneling from charged metallic grains to neutral grains, where the density of charged grains is proportional to $\exp(-E_c/kT)$. We have observed a new conduction effect in granular metals in a temperature range where the density of thermally excited charged grains is negligible, and conduction is induced by the application of a large electric field. We show that in this regime the major contribution to the conductivity is due to field-induced tunneling between neutral metal grains separated by one or more barriers.

In this Letter we report the experimental results only for granular Ni. The results for other granular metals (Au and Al) were found to be similar, and will be published subsequently. Granular Ni-SiO_2 films were made by cosputtering from a Ni and a SiO_2 target onto fused-quartz substrates by using the technique of Hanak *et al.*[5] The substrate was maintained at room temperature by water cooling. The relative volume composition x of Ni was determined from thickness measurements.[5] Electron micrography and diffraction showed the specimens to be composed of irregu-

TABLE I. Experimental and theoretical values of the granular-Ni parameters.

x (%)	d (Å)	s (Å)	E_c (theor) (meV)	σ_0 (theor) ($10^{-4}\ \Omega^{-1}\ \mathrm{cm}^{-1}$)	σ_0 (exp)	$\mathcal{E}_0$ (theor) (10^5 V cm^{-1})	$\mathcal{E}_0$ (exp)
42	66	5.2	28	2200	60	4	3
38	62	6.8	39	1300	13	7	4
32	55	9.5	64	600	13	18	8
27	47	12	101	140	5	40	21

larly shaped fcc nickel grains embedded in a silica matrix.[6] The mean grain size d and the composition x for four samples are given in Table I. The standard deviation for the grain size distribution at any composition is about 0.3. The uncertainty in x is about 15%.

To measure the conductivity, the granular metals were sandwiched between two evaporated gold films, and the voltage was applied across the thickness l of the films. The area across which the conductivity was measured was 0.1×0.025 cm^2. The current-voltage characteristics were measured by standard dc techniques. The field dependence of the conductivity σ at 1.25°K (defined as the ratio of current density j to electric field $\mathcal{E}$) for the samples given in Table I is plotted in Fig. 1. With increasing temperature, the dependence of σ on E becomes progressively weaker. Near room temperature σ is field independent and has a temperature dependence of the form $\sigma \sim \exp(-E_c/kT)$, in agreement with previous work.[2-4]

We now derive, on the basis of a simple physical model of volume generation of electron-hole pairs, the observed field dependence of the conductivity, $\sigma \sim \exp(-\mathcal{E}_0/\mathcal{E})$. The temperature is assumed to be low enough so that in the absence of an electric field all the grains are neutral. In order to transfer an electron from one neutral grain to another, the charging energy E_c must be provided by the external field. The conducting electron tunnels only between those grains whose potential difference is $\geq E_c/e$. The initial and final states associated with such a tunneling process are illustrated in Fig. 2. In order to take into account that in the granular metal there is a distribution of tunneling-barrier thicknesses, we introduce a continuous variable y and express the barrier thickness by ys and the voltage difference between grains by $y\Delta V$, where s is the average barrier thickness between adjacent grains and ΔV is the average voltage drop between adjacent grains. The condition $y\Delta Ve \geq E_c$ yields a current proportional to $\exp(-\mathrm{const}\,ys)$, where $ys = (E_c/e\Delta V)s$ is the minimum thickness of tunneling barriers the electron crosses. Included in the definition of y is the possibility of an electron tunneling through several adjacent barriers. From the above discussion it is apparent that the physical origin of the exponential dependence of the current on inverse electric field is that the effective tunneling-barrier thickness is inversely proportional to the electric field.

To calculate the current density in more detail, we must first determine the hole generation rate

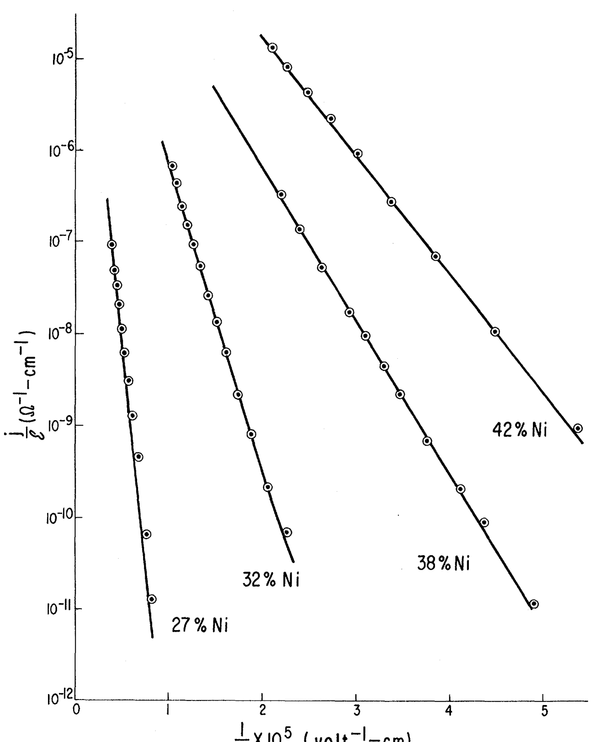

FIG. 1. Measured conductivity $j/\mathcal{E}$ at 1.25°K is plotted as a function of reciprocal field $\mathcal{E}^{-1}$. The volume composition of Ni for each sample is indicated in the figure. The experimental data, indicated by the circles, are fitted by the function $\sigma_0 \times \exp(-\mathcal{E}_0/\mathcal{E})$, indicated by the solid lines.

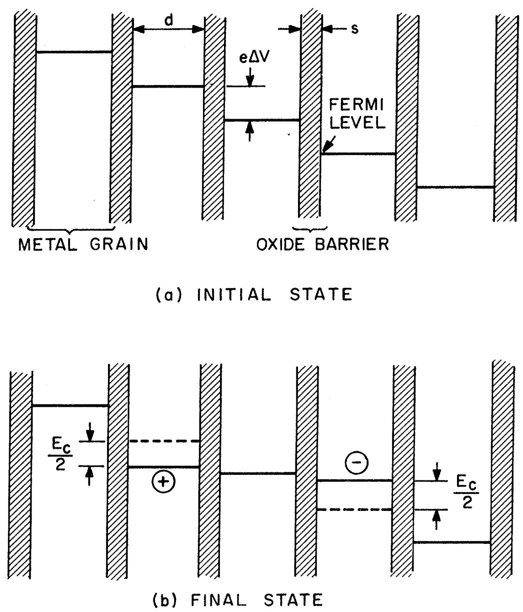

FIG. 2. Schematic representation of the effect of an electric field on the electronic energy levels in the granular metal. The voltage drop between adjacent grains is ΔV. For the sake of illustration the structure is taken as uniform (in the actual material there is a distribution of grain sizes and barrier thicknesses) and tunneling is taking place across two barriers. (a) Each grain is neutral before the tunneling process takes place. (b) After tunneling has occurred, a hole is left on one grain and an electron is added to the other grain. The relative potential levels indicated are those as seen by a negative test charge. The hole and the electron will then drift to the electrodes.

for a given grain. At $T=0$, the probability that an electron will tunnel, in unit time, through a barrier of thickness ys to another grain is given by

$$r_y = \int_0^\infty AG(v/2d)\theta(E_F - E)\theta(E - E_F + y\Delta V e - E_c) \times \exp[-2\chi(E)ys]\rho(E)\,dE. \quad (1)$$

Here G is the grain volume, v is the electron velocity, E_F is the Fermi energy, E is the electron energy, $\rho(E)$ is the density of states, and $\chi = [2m(\varphi + E_F - E)/\hbar^2]^{1/2}$, φ being the effective barrier height; the function $\theta(u)$, defined such that $\theta(u) = 1$ for $u \geq 0$ and $\theta(u) = 0$ for $u < 0$, ensures the conservation of energy. Since in our present case the condition $\varphi \gg e\Delta V$ is always satisfied, we have neglected in Eq. (1) the effect of applied field on the barrier shape. The factor $(v/2d) \times \exp(-2\chi ys)$ is the tunneling rate in the WKB approximation for the case of spherical symmetry,[7] $v/2d$ being the collision frequency of the electron with the barrier. A is a geometric factor which takes into account the fact that tunneling occurs across nonplanar barriers. For a planar barrier of uniform thickness, $A=1$. For the case of tunneling between two irregularly shaped grains, the value of A can be considerably smaller than 1 because the "effective" area for tunneling is smaller.

The current density j is given by the number of electrons and holes crossing a plane of unit area per second. Neglecting electron-hole recombination we obtain

$$j = \int_0^\infty eNlr_y\,dy, \quad (2)$$

where N is the number of metal grains per unit volume. Substituting Eq. (1) in Eq. (2) and neglecting higher-order terms in the integrals, we obtain, for the case of a system of randomly distributed spherical grains with diameter d,

$$j/\mathcal{E} = \sigma_0 \exp(-\mathcal{E}_0/\mathcal{E}), \quad (3)$$

where

$$\sigma_0 = [\rho(E_F)e^2 l v_F]/[8d\chi_0^5(d+s)^2 s^2], \quad (4)$$

and

$$\mathcal{E}_0 = 2\chi_0 s E_c/e(d+s), \quad (5)$$

with $\chi_0 = (2m\varphi/\hbar^2)^{1/2}$. The electric field $\mathcal{E}$ is defined as V/l.

To determine E_c,[2,3] we note that the energy required to remove an electron from a neutral grain to another neutral grain is $2e^2/\epsilon d$, where ϵ is the dielectric constant of the insulator. The resulting hole and electron will polarize the surrounding metal grains and the energy will be lowered by $-e^2/\epsilon(\frac{1}{2}d + s)$. The resulting E_c is therefore $2se^2/\epsilon d(\frac{1}{2}d + s)$.

In Fig. 1, the experimental points are fitted with a function of the form $\sigma_0 \exp(-\mathcal{E}_0/\mathcal{E})$. In Table I the fitted values of σ_0 and $\mathcal{E}_0$ are compared with the values calculated from Eqs. (4) and (5). For $\rho(E_F)$ and v_F we have used the values of 1.88 $\times 10^{22}$ eV^{-1} cm^{-3} and 1.6 $\times 10^{8}$ cm sec^{-1}, respectively, computed on the assumption of a free-electron mass and a free-carrier concentration n of 8.8 $\times 10^{22}$ cm^{-3} based on Hall measurements in nickel.[8] For ϵ we have used the value[9] 2.1 for SiO_2. For d we have used the values obtained from electron micrography. For the average barrier thickness s we have used the distance of closest approach between two metal spheres in a cubic lattice, with metal spheres occupying a fraction x of the total volume. The values of s

and E_c used in the calculation are given in Table I. To estimate the value of χ_0 we have used the barrier height[10] $\varphi = 3.6$ eV. In view of the simplicity of the model and the uncertainty in the experimental parameters, the agreement between theory and experiment for the quantity $\mathcal{E}_0$ is considered satisfactory (the uncertainty in the composition alone can account for the discrepancy). On the other hand, the theoretical value for the pre-exponential factor σ_0 exceeds the experimental one by a factor 30–100. Such a discrepancy is not surprising since σ_0 is very sensitive to the assumed shape and size of the grains and barriers and to the value of χ_0. The actual irregular shape of the grains could result in a substantially lower value of A than that corresponding to spherical grains. Also a variation in the value of d by a factor of 2 results in a factor of 30 for σ_0. The effects of finite energy-level separation, which for an irregular shaped 50-Å nickel grain is of the order of $E_F/nG \sim 1$ meV $\ll E_c$, have been neglected in view of the energy smearing introduced by the fluctuations in the charging energy. Such fluctuations are expected to result from electron-hole interactions. The temperature dependence of the conductivity, together with other related results, will be reported elsewhere.

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Effects of Electron-Plasmon Coupling on the Magneto-Optical Properties of Semiconductors

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Magnetotransmission experiments have been carried out on InSb samples ($4\times10^{14} < N < 6\times10^{16}$ cm^{-3}). These experiments reveal new absorption lines which are interpreted in terms of electron-plasmon interaction. Results are compared with a theoretical calculation.

In this Letter we report the first unambiguous observation of resonance absorption of infrared radiation due to plasmon-assisted electronic transitions in degenerate InSb in a magnetic field. To our knowledge this represents the first clear evidence of an absorption process which depends upon the interaction between individual conduction electrons and plasmons in semiconductors.

Electron-plasmon coupling in solids has been discussed by many authors since the pioneering