OBSERVATIONS OF SURFACE PLASMON EXCITATION BY TUNNELING ELECTRONS IN GaAs-Pb TUNNEL JUNCTIONS

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Structure in d^2I/dV^2 , which corresponds to an increase in conductance at bias voltages equivalent to the surface plasmon energy in GaAs, is observed in *n*-type GaAs-Pb tunnel junctions. Its dependence on the electron concentration of the GaAs electrode has been studied. These data are consistent with the interpretation that the observed structure in d^2I/dV^2 is due to surface plasmon excitation in the GaAs electrode by tunneling electrons.

Recent studies of tunnel junctions have shown that tunneling electrons can excite molecular and phonon excitations in the tunneling barrier.¹⁻⁴ These barrier excitation processes give rise to an increase in conductance at bias voltages equal to the excitation threshold energy. We wish to report in this Letter our observation of structure in d^2I/dV^2 for *n*-type GaAs-Pb surface-barrier tunnel junctions at bias voltages equivalent to the surface plasmon excitation energy of the GaAs electrode. This structure also corresponds to an increase in conductance in both bias directions. Its dependence on electron concentration in the GaAs electrode is consistent with the interpretation that it is due to the excitation of surface plasmons in GaAs by tunneling electrons. We believe that these preliminary results constitute the first reported observation of surface plasmon excitation in a degenerate semiconductor.⁵

The experimental procedure is similar to that described earlier.⁶ The *n*-type GaAs used in these tunnel junctions are either Te- or Se-doped single crystals. All of the junctions investigated show the proper tunneling characteristics of superconducting lead at liquid helium temperatures. When lead superconductivity is quenched by applying a magnetic field of 5 kG, the background dynamical resistance (dV/dI) of these junctions is in qualitative agreement with that observed by Conley and Mahan in *n*-type GaAs-Au junctions.⁸ The zero-bias resistance peak observed in these *n*-type GaAs-Pb junctions has a half-width of about 10 meV and magnitude $\leq 0.5\%$ of the zero-bias resistance.

Figure 1 shows the recorder traces of d^2I/dV^2 signals of two junctions whose GaAs electrodes have electron concentration of 9.5×10^{18} cm⁻³ and 5.4×10^{18} cm⁻³. The junctions are at 1°K with the lead electrode superconducting. The strong structure at bias voltages of about ±36.5 meV is due to self-energy effects from electron-longitudinal optical (LO) phonon interaction in the GaAs

electrode.^{6,8} The fine structure at 41 and 45 meV arises from a reflection of the structure in the superconducting Pb density of states at the LOphonon structure at 36.5 meV.⁶ (The main superconducting Pb density-of-states structure at low bias voltages is not shown.) The bias position of the broad peaks marked by arrows is strongly dependent on the electron concentration, n, of the GaAs electrode. It moves from $\pm 80 \text{ meV}$ for $n = 9.5 \times 10^{18} \text{ cm}^{-3}$ to $\pm 60 \text{ meV}$ for $n = 5.4 \times 10^{18}$ cm^{-3} . This structure is due to surface plasmon excitation in the *n*-type GaAs electrode. It corresponds to an increase in conductance at bias voltages equivalent to the excitation threshold energy. This slight conductance increase is also observable in curves of dV/dI versus bias voltage.

Detailed data on the measured surface plasmon threshold energy, ω_S , together with some pertinent information on the investigated *n*-type GaAs crystals, are listed in Table I. The volume plasma frequency, ω_p , of the *n*-type GaAs is calculated from $\omega_p = (4\pi ne^2/m^*\epsilon_{\infty})^{1/2}$, using the electron effective mass $m^* = 0.08m_0$ and the high-frequen-



FIG. 1. d^2I/dV^2 signal from *n*-type GaAs-Pb surface barrier tunnel junctions at 1°K. The bias voltage, whose sign refers to that of the Pb electrode, is measured from the superconducting Pb energy gap.

Electron concentration n	Electron mobility μ	$\begin{array}{c} \mathbf{Plasma} \\ \mathbf{frequency} \\ \omega \end{array}$	Measured excitation threshold energy ω	$\begin{array}{c} \text{Measured} \\ \text{half-width} \\ \Delta \omega \end{array}$	
$(10^{18} \text{ cm}^{-3})$	$(cm^2/V sec)$	p (meV)	(meV)	(meV)	$\omega_{\rm s}/\omega_{\rm s}$
0.5	1020	1 20	80	30	
5.5 6.5	1920	100	68	25	1.5
5.4	1420	90	60	20	1.5
4.2	2000	80	55	18	1.5

Table I. Data on measured surface-plasmon excitation threshold energy and pertinent information on GaAs crystals investigated.

cy dielectric constant $\epsilon_{\infty} = 11.^9$ That the value of $\omega_{\rm p}/\omega_{\rm S}$ remains constant for all the samples investigated indicates that the measured threshold energy does have a square-root dependence on the electron concentration of the GaAs electrode. If the GaAs electron plasma has a sharp boundary with a semi-infinite medium of dielectric constant equal to ϵ_{∞} , this ratio, ω_{b}/ω_{S} , should equal $\sqrt{2}$.⁵ The measured value is 1.5. This deviation from the ideal value of $\sqrt{2}$ could be partly due to the existence of a metallic lead boundary at a distance of the tunneling barrier thickness away from the plasma surface. It could also be partly due to the nonideal physical boundary between the electron plasma and the tunneling barrier. In fact, the electron density in the GaAs electrode tails from its volume value to zero at the classical turning point of the tunneling barrier in some finite distance.

The observed half-width, $\Delta \omega_{S}$, listed in Table I is the width of the surface plasmon excitation peak measured at the half-maximum of the peak. Because of the ambiguity in choosing a background, the error in these values can be as large as 20%. From our limited data, there appears to be no systematic dependence of this half-width on the electron mobility of the sample. However, the strong dependence (which appears to be linear) on the electron concentration of the GaAs sample is obvious in Table I. We suspect that the presence of the Pb film and the nonideal boundary between the electron plasma and the tunneling barrier as discussed above could also be the main source of this large half-width and its dependence on the electron concentration. The collision-broadening half-width, $\Delta \omega_{\tau}$, can be calculated from the electron mobility of the samples through $\Delta \omega_{\tau} = 1/2\tau_c$ and $\tau_c = (m^*/e)\mu$. The estimated value for these GaAs samples, using m^* =0.08, is ≤ 8 meV, which is much too small to account for the observed broad structure.

Finally, it should be noted that we have found no evidence of the volume plasmon excitation by tunneling electrons in GaAs.¹⁰ This indicates that the finite distance through which the electron density in the GaAs electrode tails to zero is greater than the electron mean free path (≲30 Å) of the samples investigated. In the bulk volume of these heavily doped crystals, the coupled plasmon-LO-phonon modes are the high-frequency plasma mode having energy $\sim \omega_D$ and the low-frequency LO-phonon mode having energy close to the TO-phonon energy (i.e., $\leq 33 \text{ meV}$).⁹ The fact that the LO-phonon structure observed in all these tunnel junctions occurs at the low electron concentration $(n < 10^{17} / \text{cm}^3)$ LO-phonon energy $(\sim 36.5 \text{ meV})$ is further evidence that the tunneling electrons never penetrate beyond the electron density tailing layer of the crystal.

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¹R. C. Jaklevic and J. Lambe, Phys. Rev. Letters <u>17</u>, 1139 (1966).

²D. J. Scalapino and S. M. Marcus, Phys. Rev. Letters 18, 459 (1967).

³J. M. Rowell, W. L. McMillan, and W. L. Feldmann, to be published.

⁴I. Giaever and H. R. Zeller, Phys. Rev. Letters <u>21</u>, 1385 (1968).

⁵Surface plasmons in metals, whose excitation energy is usually a few electron volts, have been observed by studying the characteristic energy loss of fast electrons. See, for example, C. J. Powell and J. B. Swan, Phys. Rev. <u>116</u>, 81 (1959); E. A. Stern and R. A. Ferrell, ibid. <u>120</u>, 130 (1960).

⁶D. C. Tsui, Phys. Rev. Letters 21, 994 (1968).

⁷J. M. Rowell and L. Kopf, Phys. Rev. <u>137</u>, A907 (1965).

⁸J. M. Conley and G. D. Mahan, Phys. Rev. <u>161</u>, 907 (1965).

⁹See, for example, A. Mooradian and G. B. Wright, Phys. Rev. Letters <u>16</u>, 999 (1966).

¹⁰After this manuscript was prepared, we have learned that C. B. Duke, M. J. Rice, and F. Steinrisser (to be published) have observed similar d^2I/dV^2 structure in the reverse bias of their *n*-type GaAs-In tunnel junctions. They attribute their results to self-energy ef-

fects from electron-plasmon interaction in the bulk volume GaAs electrode. We have observed d^2I/dV^2 structure in both bias directions corresponding to an increase in conductance. Our data, as discussed in the text, are consistent with our interpretation that the d^2I/dV^2 structure is due to surface plasmon emission in the surface layer of the bulk GaAs electrode.

MAGNETIC AND SEMICONDUCTING PROPERTIES OF SmB₆†

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 ${\rm SmB}_6$ has been found to show no magnetic ordering down to a temperature of 0.35° K. We interpret this to mean that the Sm ion is in its nonmagnetic divalent configuration at low temperatures and find direct evidence for the changing electronic configuration of Sm from trivalent to divalent with decreasing temperature in the observed semiconducting behavior at low temperatures.

 SmB_6 is found to change from a metal to a semiconductor with decreasing temperatures and to show intrinsic semiconducting behavior down to 3°K. The compound prepared by two different methods shows no evidence for magnetic ordering down to temperatures as low as 0.35°K. This contrast to all the other magnetic rare-earth hexaborides¹⁻³ leads to the tentative conclusion that at low temperatures, where the ordering is to be expected, the Sm ion is divalent and has the $4f^6$ configuration. In that configuration the spin and orbital angular momenta cancel, and of course no ordering is expected. As will be shown below, there is an intimate connection between the semiconducting and magnetic properties which follow directly from the change of valence of the Sm ion as a function of temperature. At and near room temperature, x-ray absorption measurements⁴ and magnetic susceptibility data⁵ indicate that the Sm is present in both its divalent and trivalent state, with the latter being most prevalent.

The cubic CsCl-type structure of the rareearth hexaborides admits only one site for the Sm ion. This means that thermal energy at room temperature must be sufficient to ionize an electron from its $4f^6$ environment. As the temperature is lowered, then, one might expect to find an exponential temperature dependence of various properties related to carrier concentration. Striking confirmation for such a model has been found in the temperature dependence of the resistivity shown in Fig. 1.

It has been well established theoretically that the boron framework in the hexaboride lattice acquires two additional electrons per six boron atoms to complete its covalent bonds.⁶ Experimental confirmation for this model has been found by Johnson and Daane⁷ who have shown that the alkaline-earth hexaborides are semiconduc-



FIG. 1. Resistance of SmB_6 as a function of temperature. Closed circles: resistance versus T; open circles: resistance versus $10^3/T$.