ZERO-BIAS TUNNEL-CONDUCTANCE MINIMA DUE TO THE EXCITATION OF COLLECTIVE MODES IN THE BARRIER

C. B. Duke, S. D. Silverstein, and Alan J. Bennett
General Electric Research and Development Center, Schenectady, New York
(Received 17 May 1967)

We evaluate the contribution to the tunnel conductance due to inelastic tunneling processes whereby the tunneling electron excites continuum modes in the barrier. For acoustical dispersion relations $\omega_{\bar{p}} = v_s p + \omega_0$ of the continuum modes, the coherent inelastic contribution is usually proportional to $(|eV| - \hbar\omega_0)^2 \theta(|eV| - \hbar\omega_0)\theta(\hbar\omega_D - |eV|)$, in which $\omega_D = v_s p_{max} + \omega_0$. Comparison with experimental data suggests that certain zero-bias minima may be attributed to phonon excitation via inelastic tunneling.

Recent studies[1,2] of tunneling in Al-oxide-metal tunnel junctions indicate that the mechanism of inelastic tunneling via the excitation of vibronic modes of molecules adsorbed in the oxide leads to increases in the tunnel conductance for bias voltages V larger than the relevant threshold energy. We consider inelastic tunneling processes in which (continuum) collective modes of energy $\hbar\omega_{\bar{p}}$ in the barrier are excited by the tunneling electron. The electron is coupled to these modes either via impurities (the analog of the Mössbauer effect[3]) or via a coherent interaction with all the appropriate atoms in the barrier (the analog of phonon excitation by coherent slow-neutron scattering[3]). Either mechanism leads to a conductance minimum at zero bias because as the bias increases, more inelastic channels open up and hence more tunneling can occur. In this Letter we show that the excitation of phonons is a mechanism capable of describing the zero-bias conductance minima in lightly doped p-n diodes[4-6] and metal-semiconductor (MS) contacts[7] as well as metal-oxide-metal (MOM) junctions. This mechanism also leads to structure at selected critical-point energies in the phonon density of states, and in this context has been tentatively identified with observed structure in Pb-I-Pb and Al-I-M junctions.[8]

We also have investigated[9] the possibility that either coherent or incoherent excitation of antiferromagnetic magnons in the barrier can account for the "giant" conductance minima[10] in Cr-oxide-Ag(Pb) junctions. For an s-d exchange coupling $J \gtrsim 0.1$ eV, we find an additional contribution to the conductance which rises from zero at zero bias to a value comparable with the background conductance at a bias equal to the maximum acoustic magnon energy (about 30 meV in Cr_2O_3). However, calculations using the bulk magnon spectrum lead to line shapes[9] $G \propto |eV|^3$, 1 meV $\lesssim |eV| \lesssim 10$ meV, whereas the experimental[10] conductance is linear near zero bias. Although modifications of the magnon spectrum and coupling in a thin film can be used to derive a linear conductance,[9] the existing experimental data are not yet sufficiently extensive to establish unambiguously that the minima are associated with magnetic oxides, much less justify refinements to a theoretical model.

The phonon spectrum in p-n and MS junctions is taken to be that of the bulk semiconductor. Table I summarizes the empirical evidence for attributing the "broad Λ"[6] conductance minimum to inelastic tunneling with the emission of TA phonons. The observed enhancement of the minimum in p-type relative to n-type MS junctions[7] can be explained[9] either by the

Table I. Comparison of TA zone-boundary phonon energy with valley-to-peak width of the zero-bias conductance minimum in lightly doped p-n diodes. References to the data are indicated. The small width of the conductance minimum in silicon-metal contacts is thought to be associated with the presence of a thin oxide layer between the metal and silicon.[a]

Semiconductor	$\hbar\omega_{TA}$ (meV)	$\Delta(eV)_{expt}$ (meV)
InP	7.8[b]	12[d]
InAs	...	5[d]
InSb	5.3[b]	4[d]
GaP	14.3[b]	15[d]
GaAs	8.7[b]	6,[d] 10[e]
GaSb	6.1[b]	7[d]
Si	17.2[b]	~1[f,a]
Ge	8.3[c]	8[a]

[a]Ref. 7.
[b]S. S. Mitra, Phys. Rev. 132, 986 (1963).
[c]B. N. Brockhouse and P. K. Iyengar, Phys. Rev. 111, 747 (1958).
[d]Ref. 4.
[e]Ref. 6.
[f]Ref. 5.

small electron-hole mass ratio in the case of electron-impurity coupling or by the degeneracy of the valence band at Γ in the case of coherent ("bulk") electron-phonon coupling. However, the disappearance of the minimum with increasing doping ($\rho_i \gtrsim 5\times10^{19}$ cm^{-3})[7,8] suggests that electron-impurity interactions provide the relevant coupling mechanism. The range $r_0 \sim \rho_i^{-1/3}$ of the charge fluctuations in the junction decreases with increasing doping ρ_i, ultimately causing them to become less effective in scattering the tunneling electrons. Detailed calculations[9] indicate that the depth of the (deformation potential) coherent-coupling minimum is ~1 % of the background conductance and usually is exceeded by the depth of the impurity-induced minimum. Furthermore, barrier-penetration effects render the linewidth of the coherent-coupling minimum smaller than $\hbar\omega_{\rm TA}$. Summarizing, the data in Table I, the data on the impurity-concentration dependence of the strength of the conductance minima, and the apparent lack of correlation between the barrier penetration factor and the width of the minima indicate that coherent (deformation potential) coupling to (LA) phonons in the junction is not the mechanism responsible for the observed minima. Therefore, we discuss in detail only the hypothesis that the "broad-Λ"[6] conductance minima in p-n and MS junctions are due to phonon emission by the tunneling electrons caused by their (incoherent) coupling to potential fluctuations associated with charged impurities in the barrier.

The calculation proceeds via four main steps. The first step consists of applying the tunneling-Hamiltonian formalism[11] to write the current as the Fourier transform of a retarded commutator. Using the Matsubara notation[12,13] we obtain

$$j(eV) = (2e/\hbar)\,\mathrm{Im}L(i\omega_n \to eV + i\delta), \tag{1a}$$

$$L(i\omega_n) = \int_0^{1/\kappa T} L(\tau)\exp(i\omega_n\tau)d\tau,\quad i\omega_n = 2\pi n\kappa T, \tag{1b}$$

in which κ is Boltzmann's constant, T is the temperature, and $L(\tau)$ is a correlation function evaluated below.

The second step consists of taking the transition amplitude[11,14,15] to be the matrix element of the (impurity) potential

$$V(\vec{r},\tau) = \sum_{n,\vec{g}} [v(g) + \vec{\sigma}\cdot\vec{S}_n(\tau)J(g)]\exp\{i\vec{g}\cdot[\vec{r}-\vec{R}_n(\tau)]\} \tag{2}$$

between one-electron eigenstates

$$\psi(\vec{r}) = \delta_{\vec{q}_\perp}(x)\exp(i\vec{q}_{\parallel}\cdot\vec{\rho})/2\pi \tag{3}$$

localized on the left ($\vec{k}$) or right ($\vec{q}$) of the junction. Taking $J=0$, the correlation function is given by

$$L(\tau) = -\sum_{n,m,\vec{g}_1,\vec{g}_2,\vec{k},\vec{q}} V([\Delta^2+g_1^2]^{1/2})V^*([\Delta^2+g_2^2]^{1/2})M(\vec{g}_1,q_\perp,k_\perp)M^*(\vec{g}_2,q_\perp,k_\perp)G(\vec{k},\tau)G(\vec{q},\tau)$$

$$\times\langle T_\tau\{\exp[i(\vec{\Delta}+\vec{g}_1)\cdot\vec{R}_n(\tau)]\exp[-i(\vec{\Delta}+\vec{g}_2)\cdot\vec{R}_m(0)]\}\rangle, \tag{4a}$$

$$\vec{\Delta} = \vec{q}_{\parallel} - \vec{k}_{\parallel};\quad \vec{\Delta}\cdot\vec{g}_i = 0, \tag{4b}$$

$$M(\vec{g},q_\perp,k_\perp) = \int e^{igx}\chi_{q_\perp}(x)\chi_{k_\perp}^*(x)dx, \tag{4c}$$

in which $G(\vec{k},\tau)$ are the one-electron Matsubara propagators.[12] Other terms in Eq. (4a), induced by the bulk electron-phonon coupling in the left- and right-hand "metal" systems, have been shown to be small[13]; so we neglect them.

The third step consists of evaluating the sums over n, m in Eq. (4). These sums are performed by expanding[3] $\vec{R}_n(\tau) = \vec{R}_n^{(0)} + \vec{u}_n(\tau)$ and separating the coherent and incoherent contributions. For a random distribution of impurities, only the incoherent contributions are nonzero to lowest order in the fractional impurity concentration[12] $c = N_i/N$, and averaging over the impurity distributions leads to a factor $(c\delta_{\vec{g}_1,\vec{g}_2})$ in Eq. (4a). The evaluation of the expectation value in Eq. (4a) yields[3]

$$L(\tau)=-c\sum_{\vec{g},\vec{k},\vec{q}}|V([\Delta^2+g^2]^{1/2})M(\vec{g},q_\perp,k_\perp)|^2$$
$$\times G(\vec{k},\tau)G(\vec{q},\tau)\exp[-Q_0(\vec{\Delta},\vec{g})+Q(\vec{\Delta},\vec{g},\tau)], \quad (5a)$$

$$Q_0(\vec{\Delta},\vec{g})=\frac{R}{N}\sum_{i,\vec{p}}\left[\frac{2\langle n_{\vec{p},i}\rangle+1}{\hbar\omega_{\vec{p},i}}\right], \quad (5b)$$

$$Q(\vec{\Delta},\vec{g},\tau)=\frac{R}{N}\sum_{i,\vec{p}}\frac{1}{\hbar\omega_{\vec{p},i}}[(2\langle n_{\vec{p},i}\rangle+1)\cosh(\omega_{\vec{p},i}\tau)$$
$$-\sinh(\omega_{\vec{p},i}\tau)\,\mathrm{sgn}(\tau)], \quad (5c)$$

$$R=\hbar^2(\Delta^2+g^2)/2M, \quad (5d)$$

where M is an "average" ionic mass.[3]

The final step in the calculation consists of evaluating the integrals in Eqs. (1b), (4c), and (5a). We proceed by expanding $\exp[Q(\tau)]=1+Q+\cdots$ and performing the $i,\vec{p}$ sums last. The constant term corresponds to direct tunneling through the impurities. Its occurrence correlates with the observation of large prephonon currents in indirect p-n diodes showing a zero-bias conductance minimum.[5] Terms $O(Q^2)$ correspond to multiple phonon emission, and near zero bias give more slowly varying contributions[16] to $j(eV)$ than the one-phonon contributions. The one-phonon term yields, at $T=0$, a "direct-emission" contribution to the conductance for a symmetric barrier:

$$G(eV)=fG_0(E_0/\hbar\omega_D)^2\{[\cosh(eV/E_0)-1]\theta(\hbar\omega_D-|eV|)+[\cosh(\hbar\omega_D/E_0)-1]\theta(|eV|-\hbar\omega_D)\} \quad (6)$$

in which G_0 is the "background" potential-tunneling conductance,[15,17] E_0 is the ratio of the WKBJ tunneling exponent to the average barrier height at the Fermi energy, $\hbar\omega_D$ is the lowest zone-boundary acoustical phonon energy (TA mode in the semiconductors[18,19]), and $\theta(x)$ is the unit impulse function. The prefactor f ranges from 10^{-3} to 1 for charged shallow impurities in semiconductor tunnel junctions. We use Gaussian impurity potentials with range $\rho_i^{-1/3}$ to estimate that in the case of MS contacts, f is proportional to $m_c^{1/2}v_0^2$ for all impurity concentrations ρ_i, and depends linearly on $\rho_i^{4/3}$ for $\rho_i\lesssim(\hbar^2/2m_cE_0)^{3/2}$ and on $\rho_i^{-2/3}$ for $\rho_i>(\hbar^2/2m_cE_0)^{3/2}$. As the average charged-impurity potential depth v_0 itself is an increasing function of the carrier mass m_c when normalized to give the shallow-impurity binding energy, f is larger for p- than n-type MS contacts, as observed experimentally.[7] E_0 can be estimated directly from the experimental conductance curves[20] and is $\gtrsim$20 meV in most of the p-n junctions[4] and MS contacts.[7] Therefore, $E_0>\hbar\omega_D$, so that Eq. (6) gives a conductance proportional to $(eV)^2$, the width of the minimum at zero bias is determined by $\hbar\omega_D$, and the correlation noted in Table I is recovered.

Changes in the conductance due to a magnetic field, $\vec{H}$, arise in this model from the dependence of both $\chi_{q_\perp}(x)$ and the impurity potential on $\vec{H}$. For $(\hbar eH/m_cc)<(E_0,\hbar^2\rho_i^{2/3}/2m_c)$, the former dependence[21] leads only to a small change in f for $\vec{H}$ normal to the plane of the junction. The model is probably inadequate to describe the case in which $\vec{H}$ is in the plane of the junction.[22]

The temperature dependence of the conductance is determined not only by the κT broadening of the conductance given in Eq. (6) but also by the stimulated-emission and phonon absorption terms in Eq. (5). The latter terms give rise to a (positive) background conductance of magnitude proportional to $(\kappa T)^2$ when $\kappa T\ll\hbar\omega_D$. The calculated temperature-dependent line shape agrees semiquantitatively with the observed[6] "broad-Λ" minimum in GaAs.

Finally, in the limit that bias-induced alterations in the barrier-penetration factor can be ignored [e.g., $E_0\gg\hbar\omega_D$ in Eq. (6)], the direct-emission contribution to the conductance at zero temperature is proportional to $\sum_{\vec{p},i}|V_{\vec{p},i}|^2\times\theta(|eV|-\hbar\omega_{\vec{p},i})$, in which $|V_{\vec{p},i}|^2$ is a measure of the electron-phonon (or magnon) coupling and differs for various coupling mechanisms [being $(R^*/N\hbar\omega_{\vec{p},i})$ for the case considered above]. In this limit, as $T\to0$, the conductance and its derivatives are direct measures of a weighted density of states for the collective modes in the barrier. This result reveals that the $(eV)^2$ conductance is a consequence of a Debye phonon spectrum and that, more generally, tunneling experiments can be used as a direct spectroscopic probe of the spectral density of the collective modes. In particular, structure due to critical points and local modes is predicted at the appropriate values of eV by both the single[23] and multiple[24] phonon (magnon) terms in the expansion of $\exp[Q(\tau)]$ in Eq. (5a).

The authors are indebted to Dr. I. Giaever for suggesting that loss mechanisms in junctions could account for the zero-bias anomalies and for discussions of his unpublished data, to Dr. J. W. Conley, both for the use of unpublished data and for pointing out the small magnitude of TA zone-boundary phonon energies, and to Dr. G. D. Mahan for discussions.

[1]R. C. Jaklevic and J. Lambe, Phys. Rev. Letters 17, 1139 (1966).

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

[3]See, e.g., C. Kittel, Quantum Theory of Solids (John Wiley & Sons, Inc., New York, 1963), Chaps. 19 and 20.

[4]R. N. Hall, J. H. Racette, and H. Ehrenreich, Phys. Rev. Letters 4, 456 (1960); R. N. Hall, in Proceedings of the International Conference on Semiconductor Physics, Prague, 1960 (Academic Press, Inc., New York, 1961), p. 193; B. M. Vul, E. J. Zavaritskaya, and N. V. Zavaritskii, Fiz. Tverd. Tela 8, 888 (1966) [translation: Soviet Phys.—Solid State 8, 710 (1966)].

[5]R. A. Logan and J. M. Rowell, Phys. Rev. Letters 13, 404 (1964).

[6]R. M. Williams and J. Shewchun, Phys. Rev. Letters 14, 824 (1965); ibid. 15, 160 (1965). The theoretical line shapes referred to below Eq. (6) in the text were compared with Fig. 1(d) in the latter reference.

[7]J. W. Conley and J. J. Tiemann, General Electric Research and Development Center Report No. 66-C-467 (to be published); J. W. Conley and G. D. Mahan, General Electric Research and Development Center Report No. 66-C-443 (unpublished); J. W. Conley, unpublished.

[8]J. M. Rowell and W. L. McMillan, Bull. Am. Phys. Soc. 12, 77 (1967).

[9]A. J. Bennett, C. B. Duke, and S. D. Silverstein, to be published.

[10]J. M. Rowell and L. Y. L. Shen, Phys. Rev. Letters 17, 15 (1966).

[11]M. H. Cohen, L. M. Falicov, and J. C. Phillips, Phys. Rev. Letters 8, 316 (1962).

[12]A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, Methods of Quantum Field Theory in Statistical Physics (Prentice-Hall Inc., Englewood Cliffs, New Jersey, 1963), Chaps. 3 and 7.

[13]G. D. Mahan and C. B. Duke, Phys. Rev. 149, 705 (1966).

[14]J. Bardeen, Phys. Rev. Letters 6, 57 (1961).

[15]W. A. Harrison, Phys. Rev. 123, 85 (1961).

[16]J. S. Langer, Phys. Rev. 124, 997 (1961).

[17]D. J. BenDaniel and C. B. Duke, Phys. Rev. 152, 683 (1966).

[18]S. S. Mitra, Phys. Rev. 132, 986 (1963).

[19]B. N. Brockhouse and P. K. Iyengar, Phys. Rev. 111, 747 (1958).

[20]J. W. Conley, C. B. Duke, G. D. Mahan, and J. J. Tiemann, Phys. Rev. 150, 466 (1966).

[21]C. B. Duke, Phys. Letters 24A, 461 (1967).

[22]H. C. Praddaude, Phys. Rev. 140, A1292 (1965); A. G. Aronov and G. E. Pikus, Zh. Eksperim. i Teor. Fiz. 51, 281 (1966) [translation: Soviet Phys.—JETP 24, 188 (1967)]; J. Zak and W. Zawadaki, Phys. Rev. 145, 536 (1966).

[23]A. A. Maradudin, E. W. Montroll, and G. H. Weiss, Solid State Phys. Suppl. 3, 1 (1963).

[24]F. A. Johnson, Prog. Semicond. 9, 181 (1965).

STATIC QUADRUPOLE MOMENT OF THE 2^+ STATE IN ^{114}Cd DETERMINED BY COULOMB EXCITATION*

G. Schilling†
Case-Western Reserve University, Cleveland, Ohio

and

R. P. Scharenberg and J. W. Tippie
Purdue University, Lafayette, Indiana

(Received 21 June 1967)

Recent measurements of the ratios of Coulomb excitation probabilities for two different projectiles, which have been interpreted in terms of the reorientation effect,[1] are unable to give verification of the mechanism involved.[2-4] We have measured the reorientation effect and thus the quadrupole moment of the 0.558-MeV 2^+ state in Cd^{114} by observing the angular distribution of 25-MeV oxygen ions inelastically scattered off Cd^{114} nuclei. The shape of the angular distribution gives conclusive evidence of the presence of the reorientation effect. The scattered ions were detected in coincidence with the de-excitation gamma radiation over an angular range from 50° to 160° in the laboratory.

The advantages of this technique are these: (1) Only a single type of projectile is required.