

Besides affecting line shape, the demagnetizing field affects the shift which arises from bulk sample paramagnetism. Such shifts already have been considered in the NMR of hydrogen in metals⁷⁻⁹ and can be the source of considerable error in Knight-shift determinations. The shift is apparent in Fig. 2 from the location of the calculated signal crossover 0.4 G upfield from "zero" where it would have been were the protons "free." The total shift of the hydrogen line relative to a doped water reference was found to be $+0.8 \pm 0.2$ G. Correcting for the calculated demagnetization shift results in a net (negative) Knight shift of 0.4 ± 0.2 G upfield from the H_2O reference, or $|\Delta H/H| = (0.8 \pm 0.4) \times 10^{-3}\%$. Zamir and Cotts¹⁰ have reported, at 4 kG, zero shift of $NbH_{0.05}$ and a negative shift of $1.6 \times 10^{-3}\%$ for all higher concentrations. The NAR 1H line saturated rather easily, which is consistent with the anticipated¹¹ proton T_1 of about 0.2 sec.

The nonresonant A-R absorption modulation signal given by,

$$\Delta\alpha = \frac{\sigma_0 B_0 H_m \sin^2\theta}{\rho v, c^2} \left(\frac{\beta^2}{1 + \beta^2} \right), \quad (2)$$

where H_m is the modulation field, was used as an absolute attenuation calibration reference¹² and served also as a standard to establish optimum spectrometer tuning at all crystal angles. With the use of this method as a sensitivity check, the hydrogen NAR was examined at 0° , 45° , and 90° . The 1H signal vanished at 0° and 90° as expected from Eq. (1) while the nonresonant A-R signal peaked at 90° as expected from Eq. (2). The maximum hydrogen NAR signal of Fig. 2 occurred at

45° . From the integrated line intensity and the A-R modulation calibration we find the absolute attenuation of the observed proton NAR signal to be $\Delta\alpha \approx 2 \times 10^{-8} \text{ cm}^{-1}$. This agrees reasonably well with the calculated value considering that the observed shape is not a Lorentzian.

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Possible Observation of Local Plasmon Modes Excited by Electrons Tunneling through Junctions

Arnold Adams, J. C. Wyss, and P. K. Hansma

Department of Physics, University of California, Santa Barbara, California 93106

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The light emitted by small-particle tunnel junctions is partially polarized. The excess light polarized perpendicular to the junctions is possibly due to the radiative decay of localized surface plasmons that are excited by tunneling electrons. Both the angular distribution and the intensity versus photon energy of this light are in agreement with recent theoretical calculations based on the excitation and radiative decay of surface plasmons in a small metal particle located above a metal film.

Recently Rendell, Scalapino, and Mühlischlegel calculated the role of local plasmon modes in light emission from small-particle tunnel junctions.¹

A peak in light intensity was predicted at a photon energy near 1.85 eV due to radiative decay of local plasmon modes of gold particles on

an oxidized aluminum film. Early experimental data² suggested a peak near that energy, but did not drop off as rapidly at low photon energies as required by the theoretical predictions. These data also showed that the light was partially, but not totally, polarized as predicted by the theory.

Here we present data that suggest that these two disagreements are related. Specifically, the excess light in the perpendicular polarization is in agreement with theoretical predictions. There is, however, an additional source of light that is not at present understood.

Gold-particle electron tunneling junctions were prepared as previously described.² An Al film, 50–100 nm thick $\times$ 3.5 mm wide, was evaporated onto a glass slide and oxidized by brief air exposure. Gold was evaporated on the entire glass slide in a 10–15-Torr argon atmosphere to form gold particles. Five conducting Au cross strips, 15 nm thick $\times$ 3.5 mm wide, were evaporated in vacuum to produce five completed junctions. Thinner Au strips were not reliably conductive over the particles. Thicker Au strips, tried only a few times, had smaller efficiencies and shorter lifetimes.

In some runs carbon-coated nickel electron-microscope grids were magnetically held next to junctions during the particle deposition. The grids were covered with evaporated Al before the particle deposition and with evaporated Au afterwards to resemble junctions. However, the Al and Au films were roughly half as thick as junction films to allow enough electron beam transmission for electron microscopy. The micrographs of these grids were similar to the one previously published [Fig. 1(b) of Ref. 2]. Typical particle diameters were 10–30 nm. Though there were some isolated particles, most were in clusters.

Since the particle coverage was of order 10%, the number of particles was of order 3×10^9 in our 10^{-5} -m² junctions. The efficiency of these junctions, as measured by a calibrated silicon diode, was in the same range as reported previously²: $(0.5\text{--}5) \times 10^{-5}$. (Typical light outputs were 0.1–10 μ W; typical power inputs were $IV = 0.02\text{--}0.2$ W.)

Figure 1 shows the angular dependence of the light emitted from the junctions. The angle θ is measured from the normal to the junction. It was varied by rotating the junction on a goniometer. The total light intensity was measured by a photomultiplier (1P28) that was covered with a metal sleeve with a small hole.

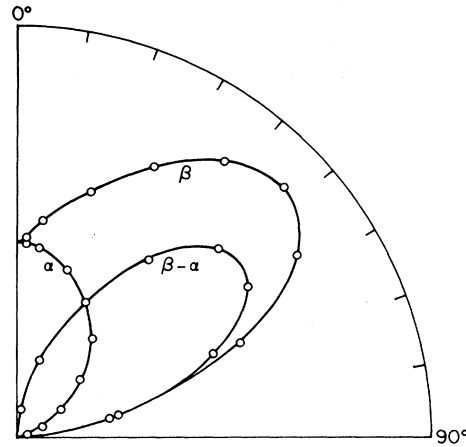


FIG. 1. The angular dependence of the light emitted from the tunnel functions at a bias voltage of 2.3 V. Component $\beta - \alpha$, the excess light in the perpendicular polarization, closely resembles the theoretical predictions for light from localized plasmon modes.

The labels α and β on the figure refer to two orientations of a Polaroid filter that was between the junction and the photomultiplier. In orientation α the transmitting axis of the filter was parallel to the plane of the junction and perpendicular to the line of observation. In orientation β the transmitting axis of the filter was in the same plane as α but rotated 90° about the line of observation.

At 0° these two components must be equal by symmetry. Away from 0° Fig. 1 shows that component β is larger. The difference between the components, $\beta - \alpha$, is the excess light polarized perpendicular to the plane of the junction. Note that this light is peaked at roughly $\theta = 55^\circ$. Its angular dependence is in agreement with recent calculations based on the theory of Rendell, Scalapino, and Mühlischlegel. In essence, it is what would be expected for a dipole radiator above a metal mirror.

Spectra were taken from 300 to 900 nm with a monochromator of ~ 2 -nm resolution and were stored on a data acquisition system. Bias voltages on the junctions varied between 1.9 and 3.3 V (current between 5 and 60 mA). Below this range of voltage and current no light was observed, and above this range the junctions would burn out quickly. Spectra were taken at different angles, θ , for both components α and β . With the use of the data acquisition system, background noise was subtracted from the data. Each spectrum was then corrected for the response of the

monochromator, photomultiplier tube, and optical components by dividing the data by the spectrum of a calibrated tungsten lamp. The resultant quotient was multiplied by the known calibrated intensity function of the lamp yielding the true light intensity of the junctions as a function of wavelength.

The results for $\theta = 0^\circ$ and 60° for a representative junction are shown in Fig. 2. At 0° component α peaks near 1.9-eV photon energy. But the peak intensity is less than 40% greater than the intensity at 1.5 eV. At 0° , as discussed above, components α and β are the same by symmetry. Thus, that the curve $\beta - \alpha$ lies at zero is a test of the two corrections for the response of the monochromator at the two input polarizations.

At 60° component α is smaller, as anticipated by Fig. 1. Its shape, however, is roughly the same as at 0° . It has a weak peak near 1.9 eV. In contrast, the excess light in the perpendicular polarization, component $\beta - \alpha$, shows a strong,

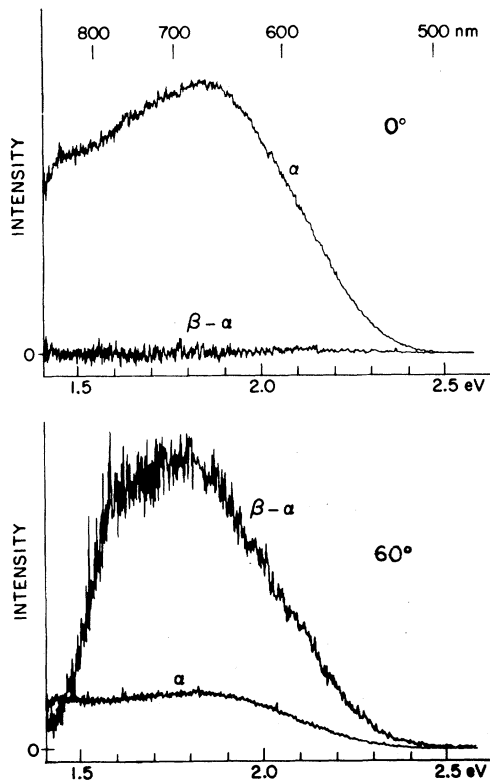


FIG. 2. Intensity (arbitrary units) versus photon energy at $\theta = 0$ and 60° relative to the normal to the junction at a bias voltage of 2.7 V. Component $\beta - \alpha$ at $\theta = 60^\circ$ closely resembles the theoretical predictions for light from localized plasmon modes.

broad peak centered near 1.85 eV. The peak intensity is roughly 600% higher than the intensity at 1.5 eV.

This strong, broad peak can be understood within the localized plasmon theory.¹ The theoretical prediction (Fig. 3 of Ref. 1) has a strong peak centered at 1.85 eV. The peak intensity is roughly 700% higher than the intensity at 1.5 eV. The only discrepancy is that the experimental peak is broader than the theoretical prediction: 0.53 eV versus 0.33 eV for the full width at half maximum. This should be expected. The theoretical prediction is for one particle. The theoretical dependence of peak position on particle diameter, d , is as $d^{-1/4}$. Thus the observed spread in particle diameters from 10 to 30 nm would correspond to a spread in peak positions of roughly $(3^{1/4} - 1) \times 1.85 \text{ eV} = 0.57 \text{ eV}$. Though more careful calculations based on observed particle distributions could be done, it is clear that the magnitude of the peak broadening is reasonable.

Thus the excess light in the perpendicular polarization agrees, both in angular dependence and in spectrum, with theoretical predictions based on a localized plasmon model. This leaves questions about the other light, component α , unanswered. Is it a measure of the unpolarized light emitted by some other mechanism? The agreement of component $\beta - \alpha$, rather than component β , with the theoretical predictions certainly suggests this. If so, what mechanism? The decay of nonlocalized plasmons in the gold film joining the particles? Scattering of emitted light by the "effective medium" of the particles? Clearly more theoretical work is needed to explain both the different angular dependence and the different spectrum of this light.

Finally, it is possible that relatively sharp peaks in light output versus photon energy that were reported for junctions with silver³⁻⁵ may be due to plasmon resonances. The form of these and their driving terms are not, however, clear at this time because of the more complex geometry.

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Exact Solution of Some Nonseparable Band Problems

Bill Sutherland

Department of Physics, University of Utah, Salt Lake City, Utah 84112

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In this paper I introduce and solve a series of band problems in higher dimensions which are generalizations of the one-dimensional Kronig-Penney model. These problems are nonseparable, in contrast to the three-dimensional model in the original Kronig-Penney work. In two dimensions the band problem is on a triangular lattice, while in three dimensions the band problem is on a body-centered-cubic lattice.

In an early and classic paper,¹ Kronig and Penney illustrated the Bloch theory of electrons in periodic potentials by a simple one-dimensional model. The potential assumed was a regular array of δ -function potentials. The resulting calculation is manageable and instructive, for it illustrates the expected band structure. Undoubtedly this simple and transparent example has helped to introduce generations of solid state physicists to an understanding of the three-dimensional band theory.

In fact, the original Kronig and Penney paper also introduced a three-dimensional band problem, by taking as a potential the sum of one-dimensional potentials in three orthogonal directions. The resulting potential then has cubic symmetry. Since this potential is separable, one easily constructs the solution from the corresponding one-dimensional solution.

In this paper I introduce and solve a series of band problems—generalizations of the Kronig-Penney model—which are not separable. The potentials are again δ functions; but in two dimensions, the lattice is a triangular lattice, while in three dimensions the lattice is bcc. To my knowledge, this is the first example of the exact solution of a nonseparable (higher-dimensional) band problem.

I wish to consider certain band problems in d dimensions,

$$-\sum_{\alpha=1}^d \frac{\partial^2 \Phi}{\partial y_\alpha^2} + V(\vec{y}) \Phi = \epsilon \Phi. \quad (1)$$

The potential V is periodic, $V(\vec{y} + \vec{\varphi}) = V(\vec{y})$, with $\vec{\varphi} = n_\alpha \vec{\varphi}_\alpha$. $\vec{\varphi}_\alpha$ are the primitive lattice vectors,

and repeated Greek indices are summed from 1 to d . The overhead arrow will always designate a d -component vector.

The particular problems I wish to consider will be derived from a corresponding periodic one-dimensional N -body system, where $N = d + 1$:

$$-\sum_{j=1}^N \frac{\partial^2 \Psi}{\partial x_j^2} + V \Psi = E \Psi. \quad (2)$$

The potential V is a sum of pair potentials,

$$V = \sum_{i>j=1}^N v(x_i - x_j), \quad (3)$$

each periodic with period unity, i.e., $v(x+n) = v(x)$ for integer n .

To exhibit the correspondence, I make a change of variables from x 's to new variables y 's, where

$$y_N = \sum_{j=1}^N x_j / \sqrt{N}$$

is the center-of-mass coordinate. I assume that the change of variables is given by an orthogonal transformation $\Lambda: y = \Lambda^{-1} x$, $x = \Lambda y$ ($\Lambda^\dagger = \Lambda^{-1}$). Note also that $\Lambda_{jN} = N^{-1/2}$. Thus

$$\sum_{j=1}^N \frac{\partial^2}{\partial x_j^2} = \frac{\partial^2}{\partial y_N^2} + \sum_{\alpha=1}^d \frac{\partial^2}{\partial y_\alpha^2}. \quad (4)$$

The particle separations are

$$\begin{aligned} x_i - x_j &= (\Lambda_{ik} - \Lambda_{jk}) y_k \\ &= (\Lambda_{i\alpha} - \Lambda_{j\alpha}) y_\alpha \equiv \lambda_{ij}^\alpha y_\alpha. \end{aligned} \quad (5)$$

(Repeated Latin indices are to be summed from 1 to N .)

One then sees that Eq. (2) separates, and I