# Raman effect in tunneling-transparent superlattices

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(Submitted 16 April 1988)
Zh. Eksp. Teor. Fiz. **94**, 251–256 (December 1988)

The cross section for the Raman scattering of light in electron superlattices is calculated, including a finite probability for tunneling between layers. In the long-wavelength approximation, i.e., for the case in which the momentum transfer is much smaller than the reciprocal of the superlattice constant, it is shown that an unscreened component of single-particle scattering arises when there is a finite tunneling. This unscreened component may be substantially greater than the scattering cross section in a lattice without tunneling. Competing "unscreened-scattering" mechanisms, unrelated to tunneling, are analyzed.

The Raman effect has recently been used to advantage in research on the electronic properties of multilayer superlattices.[1] A theory for this effect has been the subject of several papers,[2–6] but they have ignored the tunneling of electrons between layers. Since recent technological advances make it possible to synthesize very thin layers (with thicknesses as small as a few angstroms), the problem of how tunneling affects the Raman spectra in multilayer structures needs to be taken up. This is the subject of the present paper.

We know quite well that two components can be distinguished in the spectra of the Raman scattering by free carriers without spin flip: a collective component associated with plasma oscillations and a single-particle component in which the frequency transfer $\omega$ at $T=0$ is less than $\mathbf{kv}_F$, where $\mathbf{k}$ is the momentum transfer, and $\mathbf{v}_F$ the Fermi velocity. As we will show below, it is this component which undergoes the most important qualitative changes when tunneling is taken into account in the long-wavelength limit $ka \ll 1$, where $a$ is the superlatttice constant).

In the long-wavelength limit, screening effects suppress charge density fluctuations. When tunneling is taken into account, however, the electron gas in a superlattice is characterized by a very nonparabolic and isotropic dispersion law $E(p)$. In such a situation, the density of the interaction Hamiltonian of the electrons with the light wave is not proportional to the density of particles, as it is in the case of a standard band structure.[7] The result is the appearance of an unscreened component of the scattering cross section of the superlattice.

## NONRESONANT SCATTERING

If the frequencies of the incident and scattered wave, $\omega_1$ and $\omega_2$, respectively, are far from the resonances corresponding to transitions between minibands within a single band, and if $\omega_{1,2}$ are furthermore much smaller than the band gap of the host crystal, the operator representing the interaction with the electromagnetic field, described by the vector potential $\mathbf{A}$, is (see the Appendix)

$$H_{int}=\int\left[-\frac{e}{c}\Psi^{+}\frac{\partial E}{\partial p_i}\Psi A_i+\frac{e^2}{2c^2}\Psi^{+}\frac{\partial^2 E}{\partial p_i\,\partial p_j}\Psi A_iA_j\right]dV. \tag{1}$$

Here we have retained the first and second terms in the expansion in $A_i$, since only these terms are important for spontaneous Raman scattering.

We choose the following, very simple dispersion law for an electron in the superlattice:

$$E(\mathbf{p})=\frac{p_{\parallel}^2}{2m_{\parallel}}-\Delta\cos(p_{\perp}a),\quad \hbar=1, \tag{2}$$

where $p_{\perp}$ is the component of the quasimomentum along the normal to the layers. As in the case of free particles, the second-order component from the first term in (1) can be ignored to the extent that the ratios $v_F/c$ and $\Delta a/c$ are small, where $c$ is the velocity of light. With the long-wavelength limit ($ka \ll 1$) in mind, we will calculate the matrix elements $H_{\text{int}}$ in a basis of plane waves. In incorporating dynamic screening effects, we ignore flipping. We can then use the well-known procedure for calculating the scattering cross section[8,9]:

$$\frac{d^2\sigma}{d\Omega\,d\omega}=\frac{\omega^2}{\omega_1}\frac{e^4}{\pi c^4}S(\omega,\mathbf{k}), \tag{3}$$

$$S(\omega,\mathbf{k})=\operatorname{Im}\left(\Pi_2+\frac{4\pi e^2}{k^2}\frac{\Pi_1^2}{\varepsilon(\omega,\mathbf{k})}\right),$$

where

$$\varepsilon(\omega,\mathbf{k})=\varepsilon_0-\frac{4\pi e^2}{k^2}\Pi_0,$$

$$\Pi_n=\frac{2}{(2\pi)^3}\int d^3\mathbf{p}\,(\gamma(\mathbf{p}))^n\left(\frac{f_{\mathbf{p}}-f_{\mathbf{p}+\mathbf{k}}}{E(\mathbf{p})-E(\mathbf{p}+\mathbf{k})+\omega+i0}\right),$$

$$\gamma(\mathbf{p})=e_{1i}e_{2j}\frac{\partial^2 E}{\partial p_i\,\partial p_j}.$$

Here $\mathbf{e}_1$ and $\mathbf{e}_2$ are the polarizations of the incident and scattered waves, and $\varepsilon_0$ is the dielectric constant of the medium.

In the expression for the form factor $S$, it is convenient to rearrange the terms in order to single out the component which is renormalized by the Coulomb interaction of the electrons:

$$S=S_1+S_2, \tag{4}$$

$$S_1=\operatorname{Im}\frac{\Pi_1^2}{\Pi_0\varepsilon(\mathbf{k},\omega)},\qquad S_2=\operatorname{Im}\left(\Pi_2-\frac{\Pi_1^2}{\Pi_0}\right).$$

The unscreened component $S_2$ is nonzero if the electron dispersion law is not parabolic. In the case of a degenerate gas, $S_2$ is proportional to the quantity

$$\overline{(\gamma(\mathbf{p})-\overline{\gamma(\mathbf{p})})^2}|_{E=E_F},$$

where the superior bar means the average in the sense

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$$\bar{Q}=\Pi_0^{-1}\int Q(\mathbf{p})\frac{f_{\mathbf{p}}-f_{\mathbf{p}+\mathbf{k}}}{E(\mathbf{p})-E(\mathbf{p}+\mathbf{k})+\omega+i0}\frac{d^3\mathbf{p}}{4\pi^3}.$$

In multilayer superlattices an unscreened component is present only if tunneling occurs.

We assume that the barrier transparency coefficient is small enough that the width of the miniband, $\Delta$, is substantially smaller than the distance between levels in a separate layer. For the most typical electron densities, the Fermi level then lies above the top of a miniband; i.e., the Fermi surface is open in the direction away from $z$ ($\Delta \ll \pi n_s/m_\parallel$, where $n_s$ is the electron surface density). If electron scattering is ignored, the scattering cross section is nonzero in the interval $0<\omega<\omega_m$. Assuming $k_\parallel v_F \gg \Delta k_\perp a$ and $\Delta \ll E_F$, ($E_F = m_\parallel v_F^2/2 = \pi n_s/m_\parallel$), we find a simple expression for $\omega_m$: $\omega_m = k_\parallel v_F$.

In the same approximation, under the further assumptions $k\alpha$, $k\alpha_0 \ll 1$, we find

$$S_1=NS\frac{(\mathbf{e}_{1\parallel}\mathbf{e}_{2\parallel})^2}{\pi m_\parallel}\frac{\omega(\omega_m^2-\omega^2)^{1/2}}{\omega_m^2}\left(\frac{k^2aa_0}{4}\right)^2,$$
$$S_2=NS\frac{(e_{1\perp}e_{2\perp})^2}{2\pi m_\parallel}\frac{\omega}{(\omega_m^2-\omega^2)^{1/2}}\left(\frac{m_\parallel}{m_\perp}\right)^2. \qquad (5)$$

Here $S$ and $N$ are the area and number of superlattice layers, $a_0 = (m_\parallel e^2)^{-1}$ is the first Bohr radius, and $m_\perp = 1/\Delta\alpha^2$. The two components of the cross section thus differ in terms of polarization dependents, and they behave in quite different ways as $\omega$ approaches $\omega_m$. The ratio on the intergral intensities of the screened ($S_1$) and unscreened ($S_2$) components of the scattering cross section is given in order of magnitude by

$$S_1/S_2 \sim \left(\frac{k^2}{m_\parallel\Delta}\right)^2\left(\frac{a_0}{a}\right)^2. \qquad (6)$$

It follows from this estimate that even when the tunneling probability is comparatively small the unscreened component which results from the tunneling, $S_2$, may become dominant because the momentum transfer $k$ is small.

## RESONANT SCATTERING

Most experiments on the Raman effect in low-dimensionality systems are carried out under resonant conditions, under which the effect is intensified because the frequency of the incident light is approximately equal to the width of the band gap. In this case the approximation of an effective Hamiltonian (1) must be abandoned, and one should use expressions which explicitly incorporate the resonance in intermediate states in the second-order matrix element of the operator $\mathbf{A}\cdot\hat{\mathbf{v}}$ ($\hat{\mathbf{v}} = -i\nabla/m_0$, where $m_0$ is the mass of a free electron; see Ref. 9). As a result we find expressions (3), in which now the quantity $\gamma(p)$ takes the following form when only the resonant component from the subband $n'$ of band $\alpha'$ is taken into account:

$$\gamma_{\alpha n,\alpha' n'}=\frac{(\mathbf{e}_1\langle\alpha,n|\hat{\mathbf{v}}|\alpha',n'\rangle)(\langle\alpha',n'|\hat{\mathbf{v}}|\alpha,n\rangle\mathbf{e}_2)}{\varepsilon_{\alpha n}(\mathbf{p}+\mathbf{k})-\varepsilon_{\alpha' n'}(\mathbf{p})-\omega_1-i0}, \qquad (7)$$

where $\varepsilon_{\alpha n}$ and $\varepsilon_{\alpha' n'}$ are the energies of the initial and intermediate states of the crystal, respectively.

To illustrate the role played by tunneling, we consider a simple model. The wave functions between which the matrix elements are calculated in (7) are written in the strong-coupling approximation for the envelopes in the form

$$|\alpha\rangle = e^{i\mathbf{p}_\parallel\boldsymbol{\rho}}u_\alpha(\boldsymbol{\rho},z)\varphi_{\alpha p_\perp n}(z),$$
$$\varphi_{\alpha n p_\perp}(z)=\langle\varphi_{\alpha n p_\perp}|\varphi_{\alpha n p_\perp}\rangle^{-1/2}\sum_l e^{ip_\perp la}\chi_{\alpha n}(z-la), \qquad (8)$$

where $u_\alpha(\boldsymbol{\rho},z)$ is the Bloch amplitude corresponding to the extremum of band $\alpha$ at $p=0$; $x_{\alpha n}(z-la)$ is the envelope function of an electron localized in layer $l$; and $\mathbf{p}$ and $z$ are the coordinates in the plane of the layers and in the direction perpendicular to the layers, respectively.

It is assumed that only one miniband in the conduction band is populated in a superlattice of type $n$; i.e., we assume $n=0$. We will first ignore the photon momenta in the denominator of expression (7), but we will take the miniband energy, (2), into account (we assume $k_\parallel v_F \ll \Delta$). For a cubic crystal of the GaAs type, with a direct transition at the center of the Brillouin zone, we then find the following expression in the case of a resonance due to a zone which has been split off in terms of the spin ($\omega_1 \sim E_0 + \Delta_0$) (Ref. 10):

$$\gamma_{cn,vn'}=(\mathbf{e}_1\mathbf{e}_2)\left(\frac{p_{cv}}{3m_0^2}\right)^2 \times\frac{|\langle\varphi_{cnp_\perp}|\varphi_{vn'p_\perp}\rangle|^2}{E_0+\Delta_0+W_{cn}+W_{vn'}+E_{cn}(\mathbf{p})+E_{vn'}(\mathbf{p})-\omega_1}, \qquad (9)$$

where the subscripts $c$ and $v$ specify the conduction and valence bands, $W_{cn}$ and $W_{vn}$ are the energies of quantum levels for the $c$ and $v$ bands, and $E_{cn}$ and $E_{vn}$ are the kinetic energies of electrons [see (2)]. Substituting $\gamma_{cv}$ into expressions (3) and (4), we find, to leading order in the quantity $\Delta$ ($|E_g-\omega_1| \gg \Delta$) [cf. (5)],

$$S_1=NSR^2\frac{m_{\parallel e}}{\pi}\frac{\omega(\omega_m^2-\omega^2)^{1/2}}{\omega_m^2}\left(\frac{k^2a_0a}{4}\right)^2,$$
$$S_2=NSR^2\frac{m_{\parallel h}}{\pi}\frac{\omega}{(\omega_m^2-\omega^2)^{1/2}}\frac{Q_{cv}^2}{2}, \qquad (10)$$
$$R=\left(\frac{p_{cv}}{3m_0^2}\right)^2\frac{(\mathbf{e}_1\mathbf{e}_2)}{(E_g-\omega_1)}.$$

Here $E_g = E_0 + \Delta_0 + W_{cn=0} + W_{vn'} + E_F$; $\eta = 1 + m_{\parallel e}/m_{\parallel h}$; and $m_e$ and $m_{\parallel h}$ are the effective masses of the electrons and holes. The quantity $Q_{cv}$ (which is on the order of $\Delta$) is

$$Q_{cv}=A_{cv}^2\frac{(\Lambda_e m_{\parallel e}/m_{\parallel h}-\Delta_h)}{(E_g-\omega_1)}+2A_{cv}((B_{cc}+B_{vv})A_{cv}-2B_{cv}), \qquad (11)$$

where $A_{cv} = \langle\chi_{c0}(z)|\chi_{vn'}(z)\rangle$, $B_{\alpha\beta} = \langle\chi_{\alpha 0}(z)|\chi_{\beta n'} \times(z+a)\rangle$, and $\Delta_{e,h}$ are the widths of the minibands for the electrons and holes. As in (5), the unscreened component is thus proportional to $\Delta^2$, while the screened component contains the small parameter $(k^2a_0a)^2$. In principle, tunneling in terms of either of the bands, the hole band or the electron band, would be sufficient for the existence of an unscreened component $S_2$. We might add that in the resonant case the polarization dependnce corresponds to scattering by an isotropic system, in contrast with (5) (Ref. 10).

The divergences in (5) and (10) as $\omega \to \omega_m$ are actually cut off either when the finite width of the miniband is taken into account or by virtue of electron scattering. In other words, under the condition $\omega_m - \omega \lesssim \max(\delta, \tau^{-1})$ where $\tau$ is the relaxation time, and $\delta = \Delta[(k_\perp a)^2 + k_\parallel^2/2\pi n_s]^{1/2}$, the factor $\omega/(\omega_m^2 - \omega^2)^{1/2}$ assumes the value $[\omega_m/\max(\delta, \tau^{-1})]^{1/2}$.

It has thus been shown that incorporating the finite transparency of the barriers leads to an unscreened contribution to the Raman effect. There are, however, two competing contributions to the cross section $S_2$, which are not a consequence of tunneling. The first stems from the photon momenta. The quantity $\gamma$ [see (7)] depends strongly on $\mathbf{p}_\parallel$ at $E_g - \omega_1 \sim k_\parallel v_F$ (a "strong" resonance[11]). In this case an exact integration must be carried out in order to find the quantities $\Pi_n$ in (3); i.e., we do not use the expansion $f_{\mathbf{p}+\mathbf{k}} - f_{\mathbf{p}} \approx \mathbf{k}(\partial E/\partial \mathbf{p})[\partial f_0(E)/\partial E]$ (Ref. 11). In the limit $|E_g - \omega_1| \gg \omega_m$ we find

$$S_2' = NSR^2 \frac{\eta^2}{12} \frac{m_{\parallel e}}{\pi} \frac{\omega^3}{\omega_m^2(\omega_m^2 - \omega^2)^{1/2}} \frac{\omega_m^2}{(E_g - \omega_1)^2}. \qquad (12)$$

The quantity $S'_2$, associated with the strong resonance, also has a square-root singularity in the limit $\omega \to \omega_m$. Let us compare expressions (10) and (12) for the case of a small resonance defect, with $\Delta/|E_g - \omega_1| > B_{\alpha\beta}$ (or $|E_g - \omega_1| < U$, $B_{\alpha\beta} \sim \Delta/U$, where $U$ is the height of the barriers of the envelope potential of the superlattice), and the first term dominates in (11). It follows that the two contributions can be equally important, since in many experimental situations $\Delta$ is comparable to $\omega_m \approx k v_F$. In the opposite limit, $|E_g - \omega_1| > U$, the predominant mechanism at $\Delta \sim \omega_m$ is the Raman effect associated with the tunneling: $S_2'/S_2 \sim U^2/(E_g - \omega_1)^2$, $S_2'/S_2 < 1$.

Another mechanism which makes an unscreened contribution to the Raman effect is corrugation of the valence band. The dependence of the hole energy $E_v(\mathbf{p}_\parallel)$ on the direction in the $\mathbf{p}_\parallel$ plane gives rise to an angular dependence of the quantity $\gamma(\mathbf{p}_\parallel)$ [see (7)]. The corresponding contribution to the cross section, $S''_2$, vanishes in the limit $\omega \to \omega_m$. Specifically, the singular part of the quantities $\Pi_n$ is related in the limit $\omega \to \omega_m$ to the divergence of the integrals

$$\int_{-\pi}^{\pi} \frac{\omega_m \cos\varphi}{\omega - \omega_m \cos\varphi + i0} \gamma^n(\varphi)\, d\varphi, \qquad (13)$$

where we have $\omega = k v_F = \omega_m$ at the point $\varphi = 0$, at which there is a second-order pole ($\varphi$ is the angle between $\mathbf{k}_\parallel$ and $\mathbf{p}_\parallel$). Taking this circumstance into account in a calculation of $S''_2$ as $\omega \to \omega_m$, we expand $\gamma(\varphi)$ around $\varphi = 0$. It is then simple to show that we have

$$S_2''(\omega \to \omega_m) \propto (\omega_m - \omega)^{1/2}. \qquad (14)$$

A band which has been split off by the spin-orbit coupling is known to be slightly corrugated; the degree of corrugation is determined by the parameter $p_\parallel^2/m_{\parallel h}\Delta_0$. Under resonant conditions, with $\omega_1 \sim E_0 + \Delta_0$, we find the following estimate for $S''_2$: $S''_2 \sim (p_F^2/m_{\parallel h}\Delta_0)$. This estimate holds in the region $(\omega_m - \omega)/\omega_m \sim 1$).

In the experiments which have been carried out, a resonance at the frequency $\omega_1 \sim E_0 + \Delta_0$ has been used. In addition, a resonance with heavy- and light-hole bands has been used: $\omega_1 \sim E_0$. In this (less favorable) case, because of the pronounced deviation from a parabolic profile and because of the anisotropy of the hole bands, the quantity $\gamma(\mathbf{p}_\parallel)$ depends strongly on the angles in the $\mathbf{p}_\parallel$ plane and thus contributes to the unscreened cross section. At frequencies $\omega$ which are not too close to $\omega_m$, this contribution turns out to be greater than that from weak tunneling, but in the limit $\omega \to \omega_m$ we again find an $(\omega_m - \omega)^{1/2}$ law. The cross section reaches a maximum at $0 < \omega < \omega_m$. For resonances of both types $(E_0, E_0 + \Delta_0)$, the effect of the hole bands at the edge of the band, $\omega \approx \omega_m$, thus becomes small in comparison with the tunneling component.

Another unscreened process is scattering accompanied by a spin flip in the crossed-polarization geometry. In this case we can extract information about the one-particle spectrum most directly; in particular, we can determine the parameter $\Delta$. The frequency of the scattering cross section is ($k_\parallel = 0$, $k_\perp \neq 0$)

$$S \propto \omega/(\omega_m^2 - \omega^2)^{1/2}, \quad \omega_m = \Delta(k_\perp a).$$

We note in conclusion that finite barrier transparency also influences the scattering accompanied by the excitation of collective degrees of freedom, e.g., plasmons. The greatest difference from the case without tunneling arises in the situation with $k_\parallel = 0$, $k_\perp \neq 0$, of course. The form factor $S$ no longer vanishes in this case, and, as is easily shown, has the value

$$S(k_\parallel = 0, k_\perp) = R^2 \frac{k_\perp^2}{8e^2} \omega_p \delta(\omega - \omega_p). \qquad (15)$$

The frequency shift in this case is

$$\omega_1 - \omega_2 = \omega_p(k_\parallel = 0,\ k_\perp a \ll 1) = \omega_{p0}\Delta \Big/ \left(\frac{n_s 2\pi}{a^2 m_\parallel^2}\right)^{1/2},$$

where $\omega_{p0}$ is the bulk plasma frequency ($\omega_{p0}^2 = 4\pi e^2 n_s/a\varepsilon_0 m_\parallel$) (Ref. 12).

We wish to thank I. P. Ipatova and A. Maslov for useful comments and for a discussion of this study.

## APPENDIX

The scattering of light by electrons of the conduction band is described in the approximation $v_F/c \ll 1$ by the effective Hamiltonian[1,7]

$$H_{int} = \int \Psi^+ \gamma \Psi\, dV,$$

where

$$\gamma(\omega_1, \omega_2) = A_{1i}A_{2j}\left(\frac{1}{m_0} + \frac{1}{m_0^2}\sum_{\sigma \neq 0} \frac{\langle 0|\hat{p}_i|\sigma\rangle\langle\sigma|\hat{p}_j|0\rangle}{\varepsilon_0 - \varepsilon_\sigma + \omega_1} + \frac{\langle 0|\hat{p}_j|\sigma\rangle\langle\sigma|\hat{p}_i|0\rangle}{\varepsilon_0 - \varepsilon_\sigma - \omega_2}\right). \qquad (A1)$$

Here $|\sigma\rangle$ and $\varepsilon_\sigma$ are the wave functions and energies of the crystal, which incorporate the superlattice potential; $|0\rangle$ is the state of the electrons in the lower subband of the conduction band (we are assuming that only it is populated); and $m_0$ is the mass of a free electron. The energy spectrum of the superlattice is characterized by two different scale values: $E_g$, which is the band gap of the bulk crystal, and $\Omega_0$, which is distance between minibands. Under the conditions

$\omega_{1,2} \ll \Omega_0$, the applicability of (A1) is obvious. We now consider the frequency $\Omega_0 \lesssim \omega_{1,2} \ll E_g$. In this case, there is change in the contribution of only those terms which belong to the conduction band. The corresponding matrix elements are, in order of magnitude,

$$\langle 0|\hat{p}_i|\sigma\rangle \sim 1/a, \quad \Omega_0 \sim 1/m_\parallel a^2.$$

The expression used for the quantity $\gamma$ in the text proper is

$$\gamma(\omega_{1,2}=0) = \frac{\mathbf{A}_{1\parallel}\mathbf{A}_{2\parallel}}{m_\parallel} + A_{1\perp}A_{2\perp}\Delta a^2 \cos(p_\perp a); \qquad \text{(A2)}$$

i.e., it consists of an isotropic part and a part which depends on the momentum. Making use of the estimates above, we see that the corrections to each part of $\gamma$ are small quantities of the same order: $(m_\parallel/m_0)^2$. This entire discussion, of course, is valid far from resonances: $|\varepsilon_0 - \varepsilon_\sigma - \omega_2|/\Omega_0 \sim 1$.


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Translated by Dave Parsons