Intensification of multiphonon resonant Raman scattering in quasi-2D electron system

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Multiphonon resonant Raman scattering in quasi-2D electron systems (quantum wells and inversion layers) is predicted to be more intense than that in a bulk semiconductor by a large factor (α^{-1}, where $\alpha \ll 1$ is the dimensionless electron–phonon coupling constant). The threshold for this intensification shifts away from four-phonon scattering (in the volume) toward three-phonon scattering in the 2D case.

1. Multiphonon resonant Raman scattering (MRRS) has been studied experimentally and theoretically in a bulk polar semiconductor (see Refs. 1 and 2 and the bibliographies there). When a semiconductor is exposed to light with a frequency ω_l in the fundamental absorption region, one observes a series of secondary emission lines (phonon repetitions) at frequencies $\omega_s = \omega_l - N\omega_{LO}$, where ω_{LO} is the frequency of the LO phonon, and N is an integer on the order of 10 or more. The component of this multiphonon scattering which comes from free electron-hole pairs is proportional to α^3 at $N \geqslant 4$.

2. Working from the results of Ref. 3, we calculated the cross section for MRRS in the quasi-2D case. This cross section has the structure

$$\sigma = \sum_{\beta,\beta',\gamma,\gamma'} G_{\beta\beta'} S_{\beta\gamma\beta'\gamma'} f_{\gamma\gamma'} \tag{1}$$

and is determined by the fourth-rank light scattering tensor

$$S_{\beta\gamma\beta'\gamma'} = \frac{S_0}{2\pi\omega_l^2\omega_s^2} \int_{-\infty}^{\infty} dt e^{i(\omega_l - \omega_s)t} \langle \sigma^+_{\beta'\gamma'}(t)\sigma_{\beta\gamma}(0)\rangle, \tag{2}$$

$$\sigma_{\beta\gamma}(t) = -\frac{i}{\hbar S_0} \int_{-\infty}^{\infty} dt e^{i\omega\tau} \{I_\beta(t) I_\gamma(t-\tau)\}, \tag{3}$$

$$\mathbf{I} = \int dx dy \int_0^d dz \mathbf{j}(\mathbf{r}). \tag{4}$$

Here $G_{\beta\beta'}$ and $f_{\gamma\gamma'}$ are second-rank tensors associated with the geometry of the problem; the summation is over the x, y, z projections; $\langle ... \rangle$ means an expectation value over the ground state; S_0 is the normalization area; and $\mathbf{I}(t)$ is the current operator in the Heisenberg picture. The integration in (4) is over the volume of a 2D layer of thickness d. Spatial dispersion of the light has been ignored.

A calculation was carried out for a quantum well in the effective-mass approximation and in the approximation of a parabolic dispersion for the electrons and holes, by means of a diagram technique.[4] The light scattering tensor is represented as a sum over N phonon repetitions with $N \geqslant 3$:

$$S^{(N)}_{\beta\gamma\beta'\gamma'} = J_\gamma J^*_\beta J^*_{\beta'} J_{\gamma'} \frac{3^N \alpha^N \delta(\omega_l - \omega_s - N\omega_{LO})}{2\pi N^2 l^2 \omega_l^2 \omega_s^2 (\hbar\omega_{LO})^2} R_{oN}, \tag{5}$$

$$R_{oN} = \int_0^\infty dy y Z_{oN}(y) \prod_{\nu=1}^{N-1} I_\nu(y), \quad l = \left(\frac{\hbar}{2m_c \omega_{LO}}\right)^{1/2}, \quad \mathbf{J} = \frac{e}{m_0} \mathbf{p}_{cv}; \tag{6}$$

$$I_\nu(y) = (4\pi\omega_{LO}^2 l^2 / S_0) \sum_{\mathbf{k}_\nu} G_e(\mathbf{k}_\nu, \omega_\nu) G_e^*(\mathbf{k}_\nu + \mathbf{p}, \omega_\nu); \tag{7}$$

$$Z_{0N}(y) = (4\pi\omega_{LO}^2 l^2 / S_0)(\omega_0 - \omega_N)^2 \sum_{\mathbf{k}_\nu} G_e(\mathbf{k}_0, \omega_0) G_e(\mathbf{k}_0, \omega_N)$$

$$G_e^*(\mathbf{k}_0 + \mathbf{p}, \omega_0) G_e^*(\mathbf{k}_0 + \mathbf{p}, \omega_N); \tag{8}$$

$$G_e(\mathbf{k}, \omega) = [\omega - n^2 \omega_{0e} - \omega_e(\mathbf{k}) + i\gamma_n/2]^{-1}, \tag{9}$$

$$y = lp, \quad \omega_\nu = \omega_l - \omega_g - \nu\omega_{LO}, \quad \nu = 1, 2, \ldots, N.$$

Here $\mathbf{p}_{cv}$ is the interband momentum matrix element, m_0 is the mass of a free electron, $\hbar\omega_{0e} = \pi^2\hbar^2/2m_c d^2$ is the size quantization energy, n is the index of the 2D band, $\mathbf{k}_0$ and $\mathbf{k}_\nu$ are 2D wave vectors, and $\hbar\omega_g$ is the width of the band gap. The reciprocal electron lifetime γ_n in band n is determined by the electron-phonon coupling: $\gamma_n = C_n \alpha \omega_{LO}$, where $C_n \approx 1$. Analysis of expressions (6)–(8) shows that $I_1(y)$ and $Z_0N(y)$ contribute a nonresonant component of R_{0N}, while all the other $I_\nu(y)$ $(\nu = 2,..., N-1)$ are resonant and contribute a factor $\gamma_n^{-1} \sim \alpha^{-1}$. The increase in the power of α in the numerator in (5) as we go from the Nth to the $(N+1)$st phonon repetitions is thus offset by the appearance of a resonant denominator. We thus have $S^{(N \geqslant 3)}_{\beta\gamma\beta'\gamma'} \sim \alpha^2$. This result differs by a factor of α^{-1} from the result for the bulk case with $N \geqslant 4$. In other words, this theory is predicting an intensification of the phonon-repetition peaks by a large factor in cases with $\alpha \ll 1$ (in GaAs, for example, we would have $\alpha \approx 0.02$).

3. At a qualitative level, this result can be explained by arguments similar to those used in constructing a theory for MRRS in the bulk cases.[1,2] In processes of a secondary emission of light which are linear in the intensity of the exciting light, there is a spatial correlation between the electrons and the holes created by the light at one point. Once it forms, an electron-hole pair retains a finite spatial volume until it annihilates, in a process accompanied by the emission of a secondary-emission photon. The probability for this secondary emission is inversely proportional to this volume, which in turn satisfies $V_{EHP} \sim l_{ph}^3 \sim \alpha^{-3}$, where l_{ph} is the mean free path of an electron (or hole) set by the electron-phonon interaction.

In a 2D electron system, the role of this volume is played by an area, $S_{EHP} \sim l_{ph}^2 \sim \alpha^{-2}$ (a "2D volume"). Correspondingly, the probability for secondary emission is proportional to α^2, rather than α^3, and the index of the phonon repetition at which the α^2 dependence sets in correspondingly decreases from 4 to 3.

4. The effect which we have been discussing here is related to the intensification of MRRS by a factor of α^{-2} in a strong magnetic field **H** which was predicted in Ref. 4. In Ref. 4, the application of the field **H** lowered the dimensionality of the secondary-emission system. In this case the volume of an electron-hole pair is determined by the electron-phonon interaction in only a single direction: along the magnetic field. Correspondingly, the secondary-emission probability becomes proportional to α, instead of α^3, as in the absence of a magnetic field.

An intensification of MRRS in a magnetic field has recently been observed in GaAs (nine phonon repetitions) and InP (four phonon repeitions).[5]

An intensification of exciton luminescence peaks upon the application of a strong magnetic field had been observed previously in Ref. 6. The physics of that effect again involves a change in the nature of the spatial correlation of an electron and a hole when the space in which the electron-phonon coupling acts becomes quasi-one-dimensional.[4]

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