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LETTER TO THE EDITOR

The LO phonon lifetime in GaN

B K Ridley

Department of Physics, University of Essex, Colchester CO4 3SQ, UK

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Abstract. The disparity in the masses of the atomic constituents of GaN means that the usually-assumed mode of decay of a long-wavelength LO mode into two LA modes is forbidden. However, given the available information about the phonon spectrum, it appears that a possible decay route is provided by the emission of a TO and an LA phonon.

The aim here is to address the question of how an LO-phonon in GaN decays. The problem is that its zone-centre frequency is too high for the usually-assumed three-phonon process—the decay into two LA modes—to be possible. If four-phonon interactions have to be invoked the expectation would be that the lifetime would be at least an order of magnitude longer than in, for instance, GaAs, where it is a few picoseconds [1]. This would have serious consequences for electron transport in that hot-phonon effects would be very strong. The intention here is to point out that there exists a possible channel of decay that may be fast enough for the lifetime of the LO phonon in GaN to be as short as it is in GaAs. The mechanism considered is a three-phonon process whereby the LO phonon decays into a TO phonon and an LA phonon.

The original problem arises from the disparity in the masses of the two atoms of the binary compound. From recent Raman scattering experiments [2, 3] and theoretical models [4, 5] we can use the following values for the relevant zone-centre and zone-edge angular frequencies for sphalerite GaN: $\omega_{\mathrm{LO}} = 738\ \mathrm{cm}^{-1}$, $\omega_{\mathrm{TO}} = 552\ \mathrm{cm}^{-1}$, $\omega_{\mathrm{LA}} = 300\ \mathrm{cm}^{-1}$ and $\omega_{\mathrm{TA}} = 209\ \mathrm{cm}^{-1}$, where the LA and TA frequencies are the calculated zone-edge ΓX values, and the LO and TO values are from experiment. The usually assumed process cannot proceed because $2\omega_{\mathrm{LA}} < \omega_{\mathrm{LO}}$ and it is unlikely that this conclusion will be invalidated by future refinements in experiment or theory. However, the following three-phonon processes appear to be energetically allowed:

(i) $\mathrm{LO} \rightarrow \mathrm{TO} + \mathrm{LA}$
(ii) $\mathrm{LO} \rightarrow \mathrm{TO} + \mathrm{TA}$.

In either process it would be necessary for the acoustic mode to have a large wavevector in order to satisfy the energy criterion. Process (i) is energetically comfortable but process (ii) may not survive future refinements in establishing zone-edge frequencies, nor may it remain possible in all crystallographic directions. Conservation of crystal momentum in normal processes entails that the created phonons move in nearly opposite directions, because the wavevector of the LO mode of interest in transport is small. (In this note umklapp processes will not be discussed.) Symmetry considerations based on the form of a third-order process in a cubic crystal suggest for optimum effect that the polarization vectors of the three components must be mutually perpendicular (a condition which, incidentally, appears to rule out the usually assumed process of decay into two LA modes). This condition

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can be satisfied to a good approximation in process (i) if the TO mode is s-type (polarized perpendicular to the plane in which the two emitted modes lie) since the LA mode is always p-type (polarization in the plane). In process (ii) the condition cannot be satisfied strongly. We therefore focus on the interaction LO $\rightarrow$ TO(s) + LA.

Following previous approaches [6, 7, 8] we define the interaction Hamiltonian to be

$$H = \sum_{r,i,j,k,\alpha,\beta,\gamma} M_\alpha^{1/2}\omega_\alpha M_\beta^{1/2}\omega_\beta 2\Gamma_{ijk}(\alpha,\beta,\gamma)u_{i\alpha}u_{j\beta}u_{k\gamma} \tag{1}$$

where u is the relative displacement of the ions in each mode:

$$u = \sum_q \left(\frac{\overline{h}}{2NM\omega}\right)^{1/2}\left(e^* a_q^\dagger \mathrm{e}^{\mathrm{i}q.r} + e a_q \mathrm{e}^{-\mathrm{i}q.r}\right). \tag{2}$$

N is the number of unit cells, M is the oscillator mass, and e is a unit polarization vector. $\Gamma_{ijk}(\alpha,\beta,\gamma)$ is the third-order anharmonicity coefficient, the optical-mode analogue of the Grüneisen constant. The subscripts i, j, k refer to spatial directions; α, β, γ refer to the type of mode. In writing equation (1) we assume that anharmonic effects are determined by relative displacement and not by acoustic-type strains (which would expand the coupling parameter from a third-order to a sixth-order tensor). This assumption is naturally valid for optical modes and should be a reasonable approximation for short-wavelength acoustic modes.

We take the magnitude of the coupling parameter to be dependent on what modes are being coupled. For cubic crystals this coefficient is zero unless the i, j and k are all different. It is this symmetry condition that emphasises mutual perpendicularity. The net rate of annihilation of an LO phonon is given by

$$W_\gamma = \frac{2\pi}{\overline{h}}\int \frac{\Gamma^2\overline{h}^3\omega_\alpha\omega_\beta}{2NM_\gamma\omega_\gamma}(e_\alpha^* e_\beta^*)^2\delta_{\boldsymbol{q}_\gamma,\boldsymbol{q}_\alpha+\boldsymbol{q}_\beta}\left(n(\omega_\alpha)+n(\omega_\beta)+1\right)\delta(\overline{h}\omega_\alpha+\overline{h}\omega_\beta-\overline{h}\omega_\gamma)\mathrm{d}N_f \tag{3}$$

where $n(\omega)$ is the Bose–Einstein phonon occupation factor and $\boldsymbol{q}$ is the wavevector. The polarization factor can be replaced by unity without much error. We identify the LO mode with the subscript γ, the TO mode with α and the LA mode with β. To a good approximation the TO frequency can be taken to be constant across the zone and $q_\gamma \ll q_{\alpha,\beta}$. Assuming isotropy, we perform the integration over the final states by integrating over the solid angle and converting the integration over $q_\alpha (= q_\beta)$ to an integration over the LA frequency by introducing the group velocity v_{LA}. The result is

$$W_{\mathrm{LO}} = \frac{\Gamma^2\overline{h}\omega_{\mathrm{TO}}(\omega_{\mathrm{LO}}-\omega_{\mathrm{TO}})\left[n(\omega_{\mathrm{TO}})+n(\omega_{\mathrm{LO}}-\omega_{\mathrm{TO}})+1\right]q_{\mathrm{LA}}^2}{2\pi\rho_{\mathrm{LO}}\omega_{\mathrm{LO}}v_{\mathrm{LA}}} \tag{4}$$

where ρ_{LO} is the reduced-mass density, q_{LA} is the LA wavevector associated with the LA frequency $(\omega_{\mathrm{LO}}-\omega_{\mathrm{TO}})$ and v_{LA} is the group velocity.

We note that the LA frequency is 186 cm^{-1} which is small enough for the mode to lie on the part of the dispersion curve which is approximately linear. If this is assumed for the sake of simplicity, the rate for process (1) can be obtained from equation (4), thus:

$$W_{\mathrm{LO}} = \frac{\Gamma^2\overline{h}\omega_{\mathrm{TO}}(\omega_{\mathrm{LO}}-\omega_{\mathrm{TO}})^3\left[n(\omega_{\mathrm{TO}})+n(\omega_{\mathrm{LO}}-\omega_{\mathrm{TO}})+1\right]}{2\pi\rho_{\mathrm{LO}}\omega_{\mathrm{LO}}v_{\mathrm{LA}}^3}. \tag{5}$$

This has the same form as the usual expression [7]. Taking $\Gamma \approx 10^8$ cm^{-1} as for GaAs we obtain a zero-temperature rate of 5.0×10^{11} s^{-1}, which is of the same order as the observed rate in GaAs.

We conclude on the evidence available that a three-phonon process turns out to be a possible mechanism that determines the LO phonon lifetime in GaN. However, it should be pointed out that the rate, in reality, may depend crucially on the precise location of the emitted modes in the Brillouin zone. Finally, we may note that the decay route examined here is generally available in III–V compounds, including GaAs. As regards group IV materials, the assumption of a third-order coupling constant would seem to strictly rule out third-order processes owing to the presence of inversion symmetry. However, such an assumption would be carrying the approximation adopted here too far for materials where the relevant frequency gap between optical and acoustic branches is smaller than it is for polar materials, with the consequence that longer wavelength acoustic modes are involved.

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