Band in ARPES caused by photodissociation of strong-coupling large polarons

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We consider decay of phonon condensate into phonons at photodissociation of the strong-coupling large polarons when the charge carrier becomes free. Expression to calculate the band in ARPES caused by photodissociation of SCLP is obtained.

- Strong-coupling large polaron (SCLP)
- Phonon condensate in SCLP
- Decay of phonon condensate at photodissociation of SCLP
- Band in optical conductivity and in ARPES due to photodissociation of SCLP and its comparison with experiments

Electron-phonon interaction: Frohlich Hamiltonian

$$H = -\frac{\hbar^2}{2m^*}\nabla_{\mathbf{r}}^2 + \sum_{\vec{k}\neq 0}\left\{\hbar\omega_{\mathbf{k}} b_{\mathbf{k}}^{+} b_{\mathbf{k}} - \frac{e}{|\mathbf{k}|}\left(\frac{2\pi\hbar\omega_{\mathbf{k}}}{V\varepsilon^*}\right)^{1/2}\left[b_{\mathbf{k}} e^{i\mathbf{kr}} + b_{\mathbf{k}}^{+} e^{-i\mathbf{kr}}\right]\right\}$$

$$\varepsilon^* = \frac{\varepsilon_0 \varepsilon_\infty}{\varepsilon_0 - \varepsilon_\infty}$$

- Large radius (R>a) polaron occurs if the free carrier band is larger than the polaron binding energy Ep. **Continual approximation**
- Strong electron-phonon coupling occurs if Ep>> $\hbar\omega$ (phonon energy). **Adiabatic approximation**
- As a result - **strong-coupling large polaron (SCLP)**

Phonon condensate in SCLP

Strong electron-phonon interaction shifts equilibrium positions of the phonon field harmonics:

$$\langle b_k \rangle = d_k , \langle b_k^{\;+} \rangle = d_k^{\;*}$$

If $|n\rangle$ is eigenfunction of Hamiltonian of non-shifted oscillator than $U|n\rangle$ is eigenfunction of shifted Hamiltonian where

$$U = \prod_k U_k \qquad U_{\mathbf{k}} = \exp\left\{ d_{\mathbf{k}} b_{\mathbf{k}}^{+} - d_{\mathbf{k}}^{*} b_{\mathbf{k}} \right\}$$

In case of adiabatic condition applicability a trial wave function can be written in the form of product:

$$|s\rangle = \psi_0(\mathbf{r} - \mathbf{R}, \beta) \exp\left\{ \sum_{\mathbf{k} \neq 0} \left(d_{\mathbf{k}} b_{\mathbf{k}}^{+} - d_{\mathbf{k}}^{*} b_{\mathbf{k}} \right) \right\} |0\rangle$$

Finding parameters of phonon condensate with variational method

To determine $d_{\mathbf{k}} = |d_{\mathbf{k}}| e^{i\varphi_{\mathbf{k}}}$ **we find minimum of**

$\langle s|H|s\rangle - \left\langle -\frac{\hbar^2}{2m^*}\nabla_{\mathbf{r}}^2 \right\rangle$ **over** $|d_k|$ **and** φ_k **. It occurs at**

$$|d_{\mathbf{k}}| = \frac{e}{|\mathbf{k}|}\sqrt{\frac{2\pi}{V\varepsilon^*\hbar\omega_{\mathbf{k}}}}\eta_{\mathbf{k}}(\beta) \quad , \quad \varphi_{\mathbf{k}} = -\mathbf{kR} + 2\pi C(\mathbf{kR})$$

(E.N.Myasnikov et al., Phys. Rev. B 72, 224303 (2005)).

In the minimum

$$\langle s|H|s\rangle - \left\langle -\frac{\hbar^2}{2m^*}\nabla_{\mathbf{r}}^2 \right\rangle = -\sum_{\mathbf{k}\neq 0}\hbar\omega_{\mathbf{k}} d_{\mathbf{k}}^* d_{\mathbf{k}}$$

The value of β is determined by minimization of $\langle s|H|s\rangle$ **over β.**

Photodissociation of strong-coupling polaron

$$W_{if} = \frac{2\pi}{\hbar}\left|\left\langle f \left| H_{\text{int}} \right| i \right\rangle\right|^2 \delta\left(E_i - E_f\right) \quad H_{\text{int}} = \frac{e\hbar(\mathbf{kA})}{m^* c} e^{i\mathbf{Q}\,\mathbf{r}}$$

$$\left| i \right\rangle = \sqrt{\beta^3 / 7\pi}\left(1 + \beta r\right)\exp\left(-\beta r\right)\prod_{\mathbf{q}}\left| d_{\mathbf{q}} \right\rangle$$

$$\left| f \right\rangle = L^{-3/2}\exp\left(i\mathbf{kr}\right)\prod_{\mathbf{q}}\left| \nu_{\mathbf{q}} \right\rangle \qquad \hat{H}_{ph} = \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} b_{\mathbf{q}}^{+} b_{\mathbf{q}}$$

$$\delta\left(E_i - E_f\right) = \delta\left(-E_p + \hbar\Omega - \frac{\hbar^2 \mathbf{k}^2}{2m^*} - \nu\hbar\omega\right)$$

For the polarization field the photodissocaition of SCLP is quick Franck-Condon process:

$$W_{if} \propto \prod \left|\left\langle \nu_q \right| d_q \right\rangle\right|^2$$

It cannot be represented as successive radiation of single phonons by electron caused by electron-phonon interaction as it is done by A.S. Mishchenko et al., Phys. Rev. Lett. **91**, 236401 (2003), 96, 136405 (2006) since it occurs without participation of electron and electron-phonon interaction.

Probability of the electron transition into a state with the wave vector modulus **k** and direction in a body angle **dΩ**

$$dW = \frac{2\pi}{\hbar}\left\{\frac{e\hbar(\mathbf{kA})}{m^{*}c}\frac{32}{L^{3/2}}\sqrt{\frac{\pi}{7\beta^{3}}}\left(1+\beta^{-2}\left|\mathbf{Q}-\mathbf{k}\right|^{2}\right)^{-3}\right\}^{2}\prod_{\mathbf{q}}\left|\left\langle \nu_{\mathbf{q}}\middle| d_{\mathbf{q}}\right\rangle\right|^{2} d\rho(\mathbf{k})$$

$$d\rho(\mathbf{k}) = \frac{m^{*}L^{3}k(\varepsilon)}{(2\pi)^{3}\hbar^{2}}d\Omega$$

$$\hbar k(\varepsilon) = \sqrt{2m^{*}\varepsilon} = \sqrt{2m^{*}\left(\hbar\Omega - W - E_{p} - \nu\hbar\omega\right)}$$

$$dW_{\exp} = \sum_{\{\nu_q\}}^{\nu} dW \qquad \nu = \sum_{\mathbf{q}}\nu_{\mathbf{q}}$$

$$\mathbf{Q} = \mathbf{k} + \mathbf{q}_{0} \qquad \mathbf{q}_{0} = \sum_{\mathbf{q}}\mathbf{q}\,\nu_{\mathbf{q}}$$

Decay of phonon condensate

$$P_\nu = \sum_{\{\nu_{\mathbf{q}}\}}^{\nu} \prod_{\mathbf{q}} \left| \langle \nu_{\mathbf{q}} | d_{\mathbf{q}} \rangle \right|^2$$

$$\prod_{\mathbf{k}} \left| \langle \nu_{\mathbf{k}} | d_{\mathbf{k}} \rangle \right|^2 = \prod_{\mathbf{k}} (\nu_{\mathbf{k}}!)^{-1} \left| d_{\mathbf{k}} \right|^{2\nu_{\mathbf{k}}} \exp\left\{ -\left| d_{\mathbf{k}} \right|^2 \right\}$$

$$\overline{E}_{\mathbf{k}} = \sum_{\nu_{\mathbf{k}}=1}^{\infty} \hbar \omega_{\mathbf{k}} \nu_{\mathbf{k}} \left| \langle \nu_{\mathbf{k}} | d_{\mathbf{k}} \rangle \right|^2$$

$$\overline{E} = \sum_{\mathbf{k}} E_{\mathbf{k}} = \sum_{\mathbf{k}} \hbar \omega_{\mathbf{k}} d_{\mathbf{k}}^* d_{\mathbf{k}} = 2E_p$$

$$\sum_{\mathbf{k}} \left| d_{\mathbf{k}} \right|^2 = \overline{E} / \hbar\omega = 2E_p / \hbar\omega = \overline{\nu}$$

average number of phonons radiated at SCLP photoionization

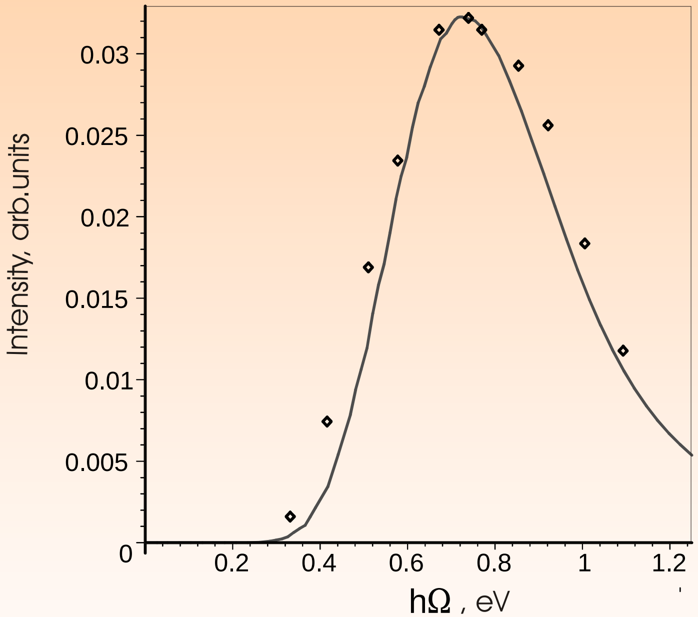
Taking into account that $d_k << 1$ the calcultaion **[E. N. Myasnikov, A.E. Myasnikova, Z.P. Mastropas, cond-mat/0703693.]** yields

$$P_\nu = \frac{\overline{\nu}^{\,\nu-1}}{(\nu-1)!} e^{-\overline{\nu}}$$

Optical conductivity

$$\mathrm{Re}\,\sigma = \frac{\hbar\Omega N_p \sum_{\nu} W(\Omega,\nu)}{\varepsilon_\infty E^2}$$

$$\mathrm{Re}\,\sigma = \frac{1024}{21}\frac{e^2 N_p}{m^*\Omega\sqrt{\varepsilon_\infty}}\sum_{\nu}\left[\varepsilon(\Omega)\frac{0.44}{E_p}\right]^{3/2}\left[1+\varepsilon(\Omega)\frac{0.44}{E_p}\right]^{-6}P_\nu$$



$$\varepsilon = \hbar\Omega - E_p - \nu\hbar\omega$$

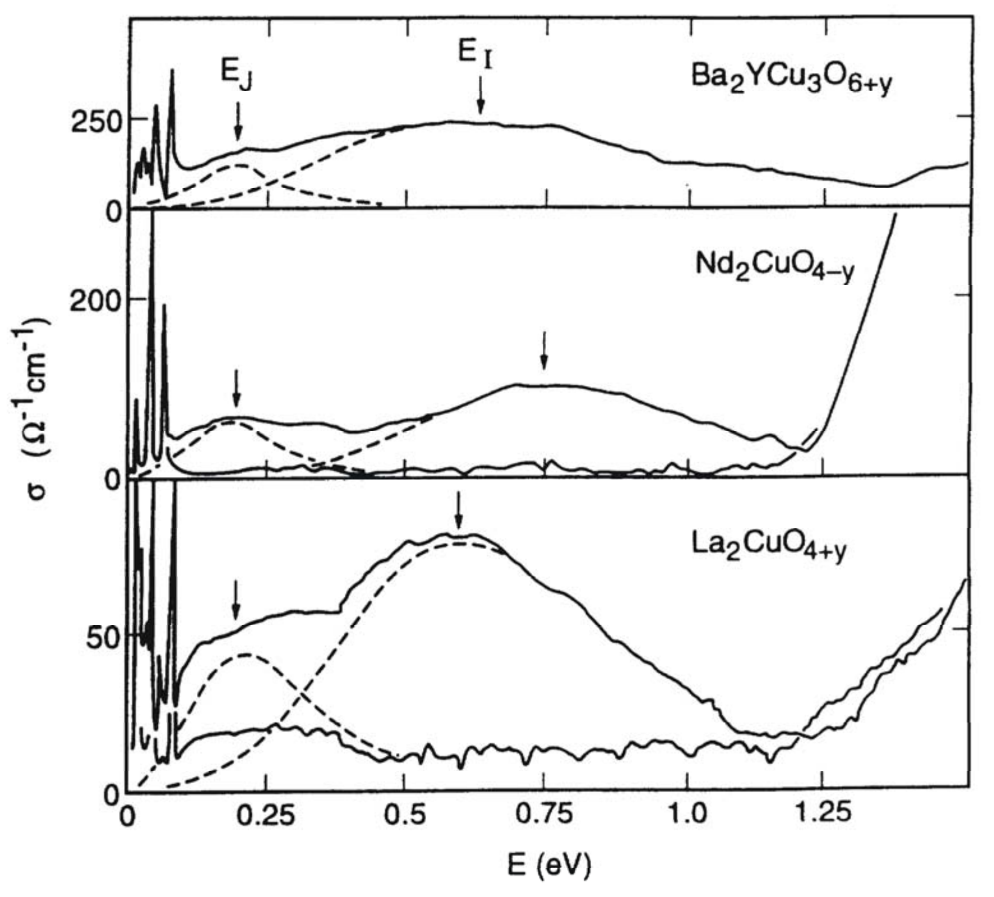
$$\hbar\Omega_{\max} = 4.2E_p$$

$$\hbar\Delta\Omega = 2.2 \div 2.8E_p$$

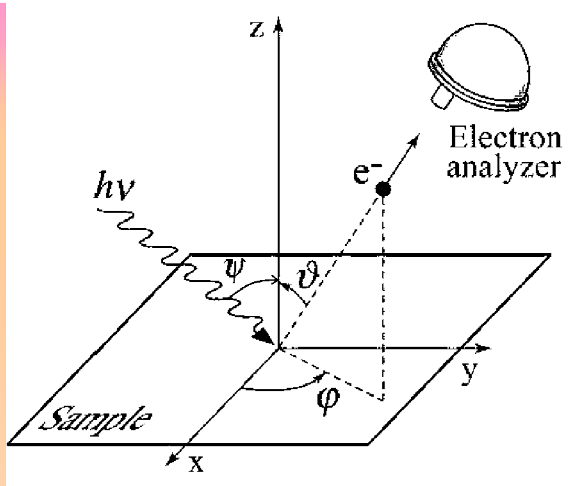
Mid-infrared band in the optical conductivity spectrum of Nd_2CuO_{4-y} : experimental, at T=10K (diamonds) and calculated with α=6, E_p =0.18 eV (solid curve).

Mid-IR bands in complex oxides

(.G. A. Thomas et al., Phys. Rev. B 45, 2474 (1992)).



ARPES from SCLP photoionization



Photoemission geometry

$$k_{\perp} = \sqrt{2m^{*}\varepsilon/\hbar^{2} - k_{||}^{2}}$$

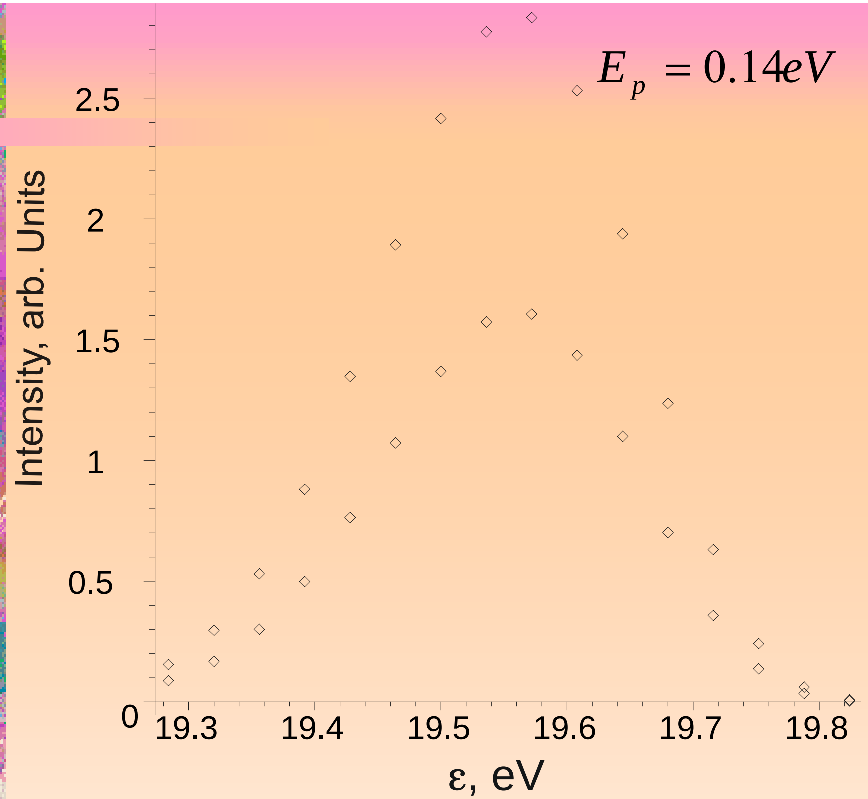
$$(\mathbf{kA}) = k_{||}\cos\varphi A\cos\psi + \sqrt{2m^{*}\varepsilon/\hbar^{2} - k_{||}^{2}}\,A\sin\psi$$

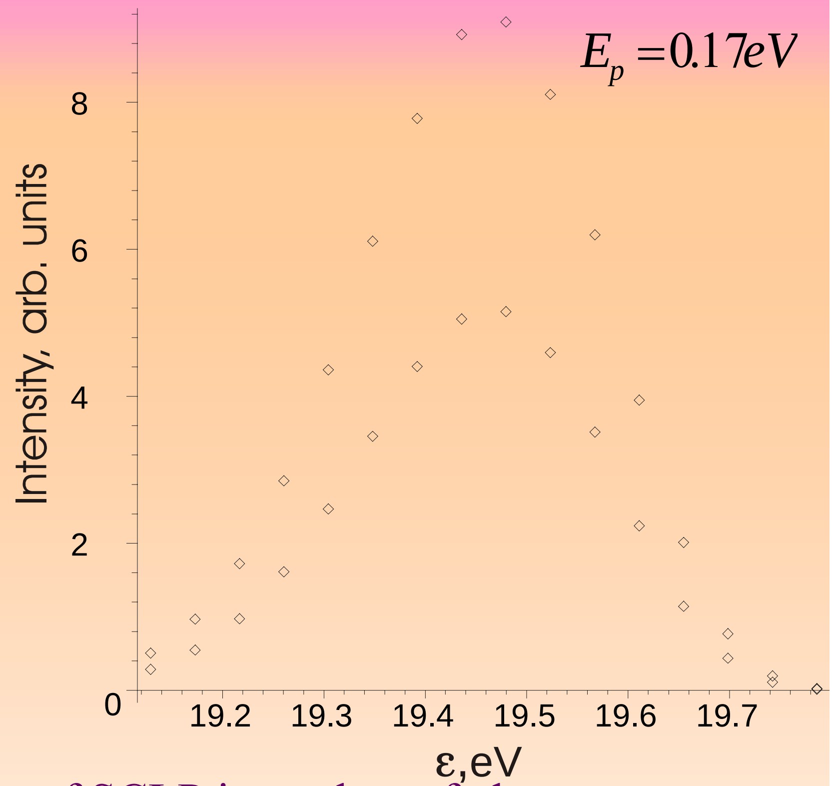
$$dW = \frac{256e^{2}}{7\pi\hbar m^{*}c^{2}\beta^{3}}\frac{(\mathbf{kA})^{2}k(\varepsilon)}{(1+\beta^{-2}k^{2})^{6}}\cdot\frac{\bar{\nu}^{\nu-1}}{(\nu-1)!}e^{-\bar{\nu}}d\Omega$$

$$\hbar k(\varepsilon) = \sqrt{2m^{*}\varepsilon} \qquad \varepsilon = \hbar\Omega - W - E_{p} - \nu\hbar\omega$$

Band maximum at $\hbar\Omega_{\max} \approx \hbar\Omega - W - 3.2E_{p}$

Band half-width $\hbar\Delta\Omega = 1.3 \div 1.7E_{p}$



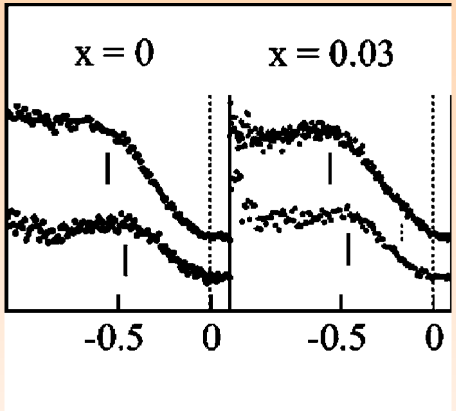


ARPES spectra caused by photoionization of SCLP in neglect of phonon dispersion, α=6, hΩ-W=20 eV. The upper and lower "curves" correspond to kx=1 and kx=0, respectively, ky=0.

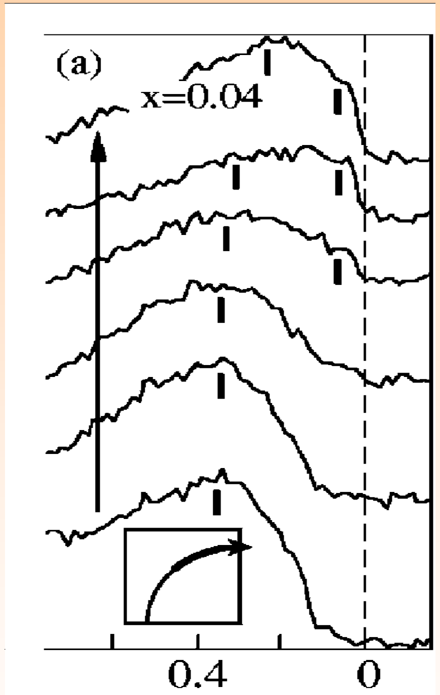
	$\hbar\Omega_{photoioniz}$, eV [14]	E_p, eV	$\hbar\Omega^{ARPES}_{max}$, eV, with respect to E_{phot}-W	$\hbar\Omega_{internal}$, eV [14]
$YBa_2Cu_3O_{6+y}$	0.62±0.05	≈0.155	≈0.48	0.16±0.03
Nd_2CuO_{4-y}	0.76±0.01	≈0.17	≈0.52	0.162±0.005
La_2CuO_{4+y}	0.6±0.02	≈0.14	≈0.44	0.13±0.02
$La_{2-x}Sr_xCuO_{4+y}$	0.53±0.05	≈0.13	≈0.4	0.16±0.03

Comparison with the experiment

ARPES spectra of underdoped $La_{2-x}Sr_xCuO_4$ and $Nd_{2-x}Ce_xCuO_4$, respectively.



(A. Ino et al., Phys. Rev. B **62**, 4137 (2000))



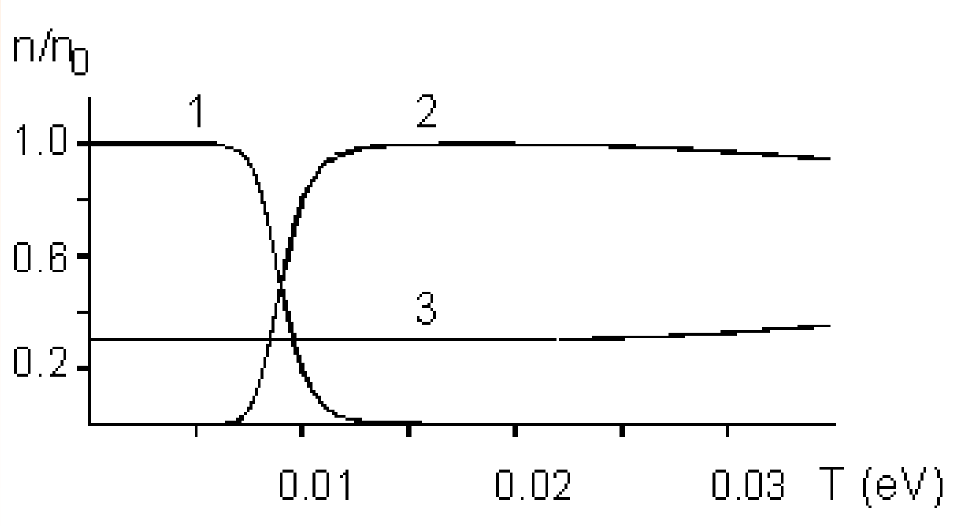
(N.P. Armitage et al., Phys. Rev. Lett. **88**, 257001 (2002))

Temperature behavior of the band caused by SCLP photodissociation

Temperature of SCLP destruction is much lower than their binding energy Ep. It can be approximated as

$$T_c = 0.278 E_p \left(\frac{m^* uc}{p_0} \right)^{0.176}$$

where m* is the bare carrier effective mass, u is the maximum group (and minimum phase) velocity of phonons interacting with the charge carrier ($\omega^2 = \omega_0{}^2 + u^2 k^2$), $c = 1/\varepsilon_0 - 1/\varepsilon_\infty$, p_0 - «maximum» momentum of the carrier in the polaron.



Accordingly, integral intensity of the band caused by SCLP photodissociation will decrease as it occurs in optical conductrivity spectra of $\beta - Na_{0.33}V_2O_5$

(C. Presura et al., Phys. Rev. Lett. 90, 026402, (2003).)