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Physica B 378–380 (2006) 267–268
PHYSICA B
www.elsevier.com/locate/physb

Phase diagram of the excitonic insulator

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Abstract

Motivated by recent experiments, which give strong evidence for an excitonic insulating phase in $TmSe_{0.45}Te_{0.55}$, we developed a scheme to quantitatively construct, for generic two-band models, the phase diagram of an excitonic insulator. As a first application of our approach, we calculated the phase diagram for an effective mass two-band model with long-range Coulomb interaction. The shielded potential approximation is used to derive a generalized gap equation controlling for positive (negative) energy gaps the transition from a semi-conducting (semi-metallic) phase to an insulating phase. Numerical results, obtained within the quasi-static approximation, show a steeple-like phase diagram in contrast to long-standing expectations.

PACS: 67.90.+z; 63.20.Kr; 65.40.−b; 64.40.Ba

Keywords: Excitonic insulator; Phase transition

The possibility of an excitonic insulator (EI) phase, separating, below a critical temperature, a semiconducting from a semi-metallic phase, has been predicted by theorists more than three decades ago [1]. However, experimental efforts to establish this phase in actual materials largely failed. It is only until recently, that detailed experimental investigations of $TmSe_{0.45}Te_{0.55}$ suggested the existence of an EI phase in this compound [2,3]. The pressure dependence of the electrical resistivity below 270 K, for instance, strongly points towards an emerging EI phase [2]. Further evidence for collective behavior which may have its origin in an EI phase comes from the linear increase of the thermal conductance and diffusivity at very low temperatures [3].

Under the assumption that the external pressure controls the energy gap E_g, the resistivity data have been used to construct a phase diagram for $TmSe_{0.45}Te_{0.55}$ in the E_g–T plane [2]. Although experimental data strongly suggest that this phase diagram is the phase diagram of an EI, to unambiguously decide if this interpretation is correct requires further theoretical examination, taking the relevant parts of the electronic structure of the material into account. However, even for the simplest two-band models, a quantitative phase diagram for an EI has never been calculated. As a first step towards a theoretical scrutiny of the phases of the Tm[Se,Te] system it is therefore appropriate to present here such a calculation.

In close analogy to the strong-coupling theory of superconductivity, we employed a matrix propagator formalism. Within a two-band model, the anomalous or off-diagonal (in the band indices $i=1,2$) self-energy $\Sigma_{12}(\mathbf{k}, i\omega_n)$ describing the pairing between conduction and valence band electrons serves as an order parameter: $\Sigma_{12}(\mathbf{k}, i\omega_n)\neq 0$ signals the existence of the EI phase. Our selfconsistent approximation enables us to take a variety of physical processes into account and results in a nonlinear functional equation for $\Sigma_{12}(\mathbf{k}, i\omega_n)$. Linearizing this equation in the vicinity of the phase boundary, where $\Sigma_{12}(\mathbf{k}, i\omega_n)$ is small, yields a generalized "gap equation". The phase boundary $T_c(E_g)$ can then be found by mapping out the T–E_g range for which the "gap equation" has nontrivial solutions.

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doi:10.1016/j.physb.2006.01.099

We applied this scheme to an isotropic, effective mass two-band model for valence and conduction band electrons interacting via the long-range Coulomb potential $V_0(\mathbf{q}) = 4\pi e^2/\varepsilon_0 q^2$. The energy gap E_g is indirect (Γ-X) and can be positive or negative. Within the shielded potential approximation the generalized gap equation for the real part of the interband self-energy reads

$$\Delta(\mathbf{k}, \tilde{e}_i) = \int \frac{d\mathbf{k}'}{(2\pi)^3}[V(\mathbf{k}-\mathbf{k}', \tilde{e}_i, \tilde{e}'_1)B_2(\tilde{e}'_2, \tilde{e}'_1)\Delta(\mathbf{k}', \tilde{e}'_1) + V(\mathbf{k}-\mathbf{k}', \tilde{e}_i, \tilde{e}'_2)B_1(\tilde{e}'_1, \tilde{e}'_2)\Delta(\mathbf{k}', \tilde{e}'_2)], \quad (1)$$

where $\tilde{e}_i(\mathbf{k}) = e_i(\mathbf{k}) - \mu_i$, $B_i(x,y) = (x-y)/[(x-y)^2 + \gamma_i(\mathbf{k})^2]$, and $V(\mathbf{k}-\mathbf{k}', \tilde{e}_i, \tilde{e}'_j) = \mathrm{Re}V^r_s(\mathbf{k}-\mathbf{k}', \tilde{e}_i - \tilde{e}'_j)n_F(\tilde{e}'_j) - \mathrm{P}\int(d\omega/\pi)1/(\tilde{e}_i - \tilde{e}'_j - \omega)\mathrm{Im}\, V^r_s(\mathbf{k}-\mathbf{k}', \tilde{e}_i - \tilde{e}'_j)n_B(-\omega)$ with $V^r_s(\mathbf{q}) = V_0(\mathbf{q})/\varepsilon^r(\mathbf{q}, \omega)$ the dynamically screened Coulomb potential. To derive Eq. (1) we employed a quasi-particle approximation for the intraband propagators with renormalized band dispersions and lifetimes given by $e_i(\mathbf{k}) = \varepsilon_i(\mathbf{k}) + \mathrm{Re}\Sigma^r_{ii}(\mathbf{k}, e_i(\mathbf{k}) - \mu_i)$ and $\gamma_i(\mathbf{k}) = -\mathrm{Im}\,\Sigma^r_{ii}(\mathbf{k}, e_i(\mathbf{k}) - \mu_i)$, respectively. The chemical potentials μ_i are measured from the respective band extrema.

The full analysis of Eq. (1) is the subject of a forthcoming publication [4]. Here, we focus on the quasi-static approximation, $V^r_s(\mathbf{k}-\mathbf{k}', \tilde{e}_i - \tilde{e}'_j) \approx V^r_s(\mathbf{k}-\mathbf{k}', 0)$, which simplifies the gap equation enormously. Except for very small band overlaps (very small Fermi surfaces), we expect this approximation to work reasonably well, as it does for intraband self-energies. For equal band masses and temperature independent screening, the quasi-static approximation reduces Eq. (1) to

$$\Delta(u) = \int_{u_1}^{\infty} du' V(u,u')\frac{\tanh u'}{2u'}\Delta(u'), \quad (2)$$

$$V(u,u') = \sqrt{\frac{1}{4\pi^2 k_B T}}\frac{1}{k}\log\left[\frac{(k+k')^2 + \kappa^2}{(k-k')^2 + \kappa^2}\right] \quad (3)$$

with $k = \sqrt{u - u_1}$, $k' = \sqrt{u' - u_1}$, $u_1 = E_g/4k_BT$, and $\kappa^2 = (2\sqrt{|E_g|}/\pi k_B T)\theta(-E_g)$. To construct the phase boundary $T_c(E_g)$, we discretize Eq. (2) and determine, for fixed E_g, the temperature $T = T_c$ for which the determinant of the coefficient matrix of the resulting system of linear equations vanishes.. For $E_g < 0$ this approach can be directly applied, whereas for $E_g > 0$, the logarithmic singularity of the kernel has to be removed first [4].

The phase boundary $T_c(E_g)$ is presented in Fig. 1, measuring energy and temperatures in units of the exciton Rydberg R_0. Above $T_1 \approx 0.45$, the EI phase is unstable. Below T_1, we find a steeple-like phase boundary which strongly discriminates between $E_g > 0$ and $E_g < 0$. For $E_g > 0$, $T_c(E_g)$ smoothly decreases to zero at $E_g = 1$, the critical band gap, above which the EI phase cannot exist. For $E_g < 0$, in contrast, $T_c(E_g)$ initially drops extremely fast, within a few percent of R_0, to a second critical temperature $T_2 \approx 0.04$. Below T_2 the EI phase is stable with an almost constant $T_c(|E_g|)$, which, however, for larger band overlaps slowly decreases (see inset). The steeple-like shape of the phase diagram reflects the different phases from which the EI is approached: semi-conducting for $E_g > 0$ and semi-metallic for $E_g < 0$. Entering the EI phase from the semi-conductor side leads to formation of *strongly bound* excitons. On the other hand, when the EI phase is approached from the semi-metal, exciton formation is strongly suppressed due to the free carrier's screening of the Coulomb potential. In that case, *loosely bound* Cooper-type pairs emerge, resulting in a rather fragile EI phase. The crossover from excitons to Cooper-type pairs occurs at $E_g \approx -0.3$, the band overlap, where the screening length becomes roughly equal to the exciton radius. For $|E_g| \ll 4k_BT$, the critical temperature is exponentially small and approximately given by $k_BT_c \approx (\gamma|E_g|/\pi)\exp(-\pi\sqrt{|E_g|}/\ln(1 + \pi\sqrt{|E_g|}/2))$,with $\gamma = \exp(0.577)$ (dashed line in the inset). Anisotropies in the band structure and other pair breaking effects would easily destroy this part of the phase diagram.

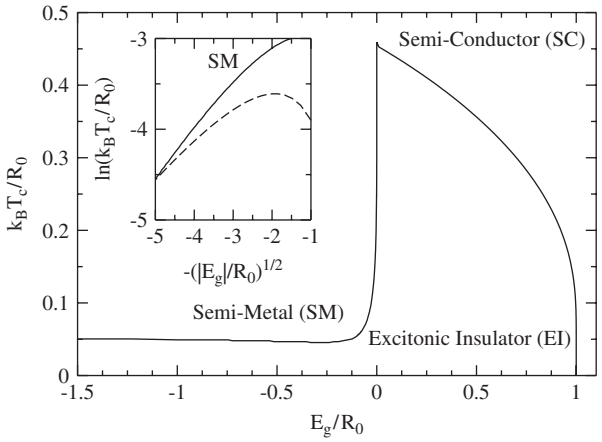

Fig. 1. Phase diagram for an excitonic insulator with equal band masses.

Support from SFB 652 is greatly acknowledged.

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