Off-diagonal disorder and metal–dielectric transition in a narrow-band metal

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The effect of impurities on the temperature T_c of the transition and on the dielectric gap is examined with a disordered binary alloy $A_{1-x}B_x$ ($x \ll 1$) as an example, with account taken of the diagonal and off-diagonal disorders. The diagonal disorder leads to the same decrease of T_c as in a superconductor with paramagnetic impurities. The off-diagonal disorder can either lower or raise T.

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1. INTRODUCTION

Consider a model of a narrow-band metal whose electron spectrum has in the tight-binding approximation the property

$$\varepsilon(\mathbf{k}+\mathbf{Q})-\mu=-\varepsilon(\mathbf{k})+\mu, \tag{1}$$

with a vector $2\mathbf{Q}$ that coincides with one of the reciprocal-lattice vectors. As shown by Kopaev and Timerov,[1] such a metal is unstable to transition into a dielectric state characterized by a gap $\Delta = g(\mathbf{Q})\langle b_{\mathbf{Q}} + b_{-\mathbf{Q}}^{+}\rangle$ (only singlet pairing is considered, $b_{\mathbf{Q}}$ is the operator of annihilation of a phonon with a wave vector $\mathbf{Q}$ and frequency $\omega_{\mathbf{Q}}$, and $g(\mathbf{Q})$ is the electron-phonon interaction constant).

From the formal point of view, the theory of Kopaev and Timerov[1] is close to the theory of an excitonic dielectric.[2] For an excitonic dielectric, the influence of the impurities was considered by Zittartz,[3] who showed that the transition temperature T_c decreases with increasing impurity concentration x in exactly the same manner as in a superconductor with paramagnetic impurities.[4] It was assumed in the Zittartz paper[3] as well as by most subsequent publications on this topic, that the impurity produces only a scattering potential and does not affect the band structure. This assumption does not hold in the case frequenctly investigated in experiments, that of isostructural substitution, i.e. for a disordered $A_{1-x}B_x$ alloy, whose components A and B have identical structures. The impurity changes here not only the charge of the ion, but the integral of the hopping to the next atom, and this changes the electron spectrum.

In the present paper we consider the metal–dielectric transition in such a model at a low concentration $x \ll 1$ of the B component, when a perturbation theory in the parameter $a/l \ll 1$ can be constructed (a is the interatomic distance and l is the mean free path). It is shown that scattering by the impurity potential (diagonal disorder) leads, just as in Refs. 3 and 4, to a lowering of T_c, whereas the fluctuations of the hopping integral (off-diagonal disorder) can both lower and raise T_c.

2. HAMILTONIAN AND GREEN'S FUNCTION

We consider a system of electrons in the tight-binding approximation

$$\mathcal{H} = \sum_{f\sigma}(\varepsilon_f-\mu)a_{f\sigma}^{+}a_{f\sigma} + \sum_{fh\sigma} b_{f,f+h}a_{f\sigma}^{+}a_{f+h\sigma} + \sum_{ff'\sigma} p_f u(\mathbf{f}-\mathbf{f}')a_{f\sigma}^{+}a_{f'\sigma}. \tag{2}$$

Here $a_{f\sigma}$ is the operator of electron annihilation in a state with a Wannier function localized on a lattice site $\mathbf{R}_f \equiv \mathbf{f}$ with spin projection σ; ε_f is a one-electron energy level in the atom $\mathbf{f}$; μ is the chemical potential; $b_{f,f+h}$ is the integral of hopping between nearest neighbors; $u(\mathbf{f}-\mathbf{f}')$ is the impurity potential; $p_f = 1$ and $p_f = 0$ respectively if an impurity is present at the site $\mathbf{f}$ or not. Taking into account the fluctuations of the energy levels and of the hopping integrals, we can write

$$\varepsilon_f = \varepsilon + p_f\delta\varepsilon, \quad b_{f,f+h} = b(\mathbf{h}) + p_f\delta b(\mathbf{h}),$$

so that after taking the Fourier transform we get

$$\mathcal{H} = \sum_{\mathbf{k}\sigma}'(\varepsilon+\xi(\mathbf{k})-\mu)a_{\mathbf{k}\sigma}^{+}a_{\mathbf{k}\sigma} + \sum_{fkk'\sigma} p_f \exp[-i\mathbf{f}(\mathbf{k}-\mathbf{k}')]v(\mathbf{k},\mathbf{k}')a_{\mathbf{k}\sigma}^{+}a_{\mathbf{k}'\sigma},$$

$$\xi(\mathbf{k}) = \sum_{\mathbf{h}} b(\mathbf{h})e^{i\mathbf{k}\mathbf{h}}, \quad v(\mathbf{k},\mathbf{k}') = \delta\varepsilon + \frac{1}{2}\sum_{\mathbf{h}}\delta b(\mathbf{h})(e^{i\mathbf{k}\mathbf{h}}+e^{-i\mathbf{k}'\mathbf{h}}) + u(\mathbf{k}-\mathbf{k}'). \tag{3}$$

We see that the level fluctuations $\delta\varepsilon$ lead simply to an increment of the Fourier transform of the impurity potential $u(\mathbf{k}-\mathbf{k}')$. The reason is that both the first and the third terms of (2) contain diagonal disorder, while the second term of (2) contains off-diagonal disorder and leads to an explicit dependence of $v(\mathbf{k}, \mathbf{k}')$ on $\mathbf{k}$ and $\mathbf{k}'$. It is convenient to separate the contributions of the diagonal and off-diagonal disorders

$$v(\mathbf{k}, \mathbf{k}') = \tilde{u}(\mathbf{k}-\mathbf{k}') + b(\mathbf{k}, \mathbf{k}'), \quad \tilde{u}(\mathbf{k}-\mathbf{k}') = \delta\varepsilon + u(\mathbf{k}-\mathbf{k}'). \tag{4}$$

If the hopping integral is the same for all neighbors, then

$$b(\mathbf{k},\mathbf{k}') = \frac{\delta b}{b}[\xi(\mathbf{k})+\xi(\mathbf{k}')]. \tag{5}$$

We consider hereafter a half-filled band, so that $\mu = \varepsilon$.

In the dielectric phase, Bose condensation of the phonons reduces the electron-phonon interaction to an external field that acts on the electrons,[1,5] and we therefore add to (3)

$$\mathcal{H}_{\Delta} = \Delta\sum_{\mathbf{k}\sigma} a_{\mathbf{k}\sigma}^{+}a_{\mathbf{k}-\mathbf{Q}\sigma}.$$

Since the lattice distortion takes place with doubling of the period, it is convenient to change over to a decreased Brillouin zone and work with matrix Green's functions. We introduce the vector column (here $\mathbf{k}$ is defined in half the initial Brillouin zone)

$$\psi_{\mathbf{k}\sigma} = \begin{pmatrix} a_{\mathbf{k}\sigma} \\ a_{\mathbf{k}-\mathbf{Q}\sigma} \end{pmatrix}, \quad \psi_{\mathbf{k}\sigma}^{+} = (a_{\mathbf{k}\sigma}^{+}, a_{\mathbf{k}-\mathbf{Q}\sigma}^{+})$$

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and the matrix Green's function in the temperature technique[6]

$$\hat{G}_\sigma(\mathbf{k}, i\varepsilon_n) = \begin{pmatrix} \langle\langle a_{\mathbf{k}\sigma} | a_{\mathbf{k}\sigma}^+ \rangle\rangle & \langle\langle a_{\mathbf{k}\sigma} | a_{\mathbf{k}-\mathbf{Q}\sigma}^+ \rangle\rangle \\ \langle\langle a_{\mathbf{k}-\mathbf{Q}\sigma} | a_{\mathbf{k}\sigma}^+ \rangle\rangle & \langle\langle a_{\mathbf{k}-\mathbf{Q}\sigma} | a_{\mathbf{k}-\mathbf{Q}\sigma}^+ \rangle\rangle \end{pmatrix} = \begin{pmatrix} G_\sigma(\mathbf{k}, i\varepsilon_n) & F_\sigma(\mathbf{k}, i\varepsilon_n) \\ F_\sigma^+(\mathbf{k}, i\varepsilon_n) & G_\sigma(\mathbf{k}-\mathbf{Q}, i\varepsilon_n) \end{pmatrix}.$$

The matrix form of the Hamiltonian is

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_1, \quad \hat{\mathcal{H}}_0 = \sum_{\mathbf{k}\sigma} \psi_{\mathbf{k}\sigma}^+ \hat{\omega}_{\mathbf{k}} \psi_{\mathbf{k}\sigma}, \quad \hat{\mathcal{H}}_1 = \sum_{\mathbf{k}\mathbf{k}'\sigma} \psi_{\mathbf{k}\sigma}^+ \hat{V}_{\mathbf{k}\mathbf{k}'} \psi_{\mathbf{k}'\sigma},$$

$$\hat{\omega}_{\mathbf{k}} = \begin{pmatrix} \xi(\mathbf{k}) & \Delta \\ \Delta & \xi(\mathbf{k}-\mathbf{Q}) \end{pmatrix}, \quad \hat{V}_{\mathbf{k}\mathbf{k}'} = \begin{pmatrix} V_{\mathbf{k}\mathbf{k}'} & V_{\mathbf{k},\mathbf{k}'-\mathbf{Q}} \\ V_{\mathbf{k}-\mathbf{Q},\mathbf{k}'} & V_{\mathbf{k}-\mathbf{Q},\mathbf{k}'-\mathbf{Q}} \end{pmatrix},$$

where

$$V_{\mathbf{k}\mathbf{k}'} = \sum_t p_t e^{-i\mathbf{t}(\mathbf{k}-\mathbf{k}')} v(\mathbf{k}, \mathbf{k}').$$

The Hamiltonian $\mathcal{H}_0$ describes a regular crystal in the dielectric state, and the corresponding Green's function $\hat{G}_0(\mathbf{k}, i\varepsilon_n)$, where $\varepsilon_n = (2n+1)\pi T$, is given by

$$\hat{G}_0^{-1}(\mathbf{k}, i\varepsilon_n) = i\varepsilon_n \hat{I} - \hat{\omega}_{\mathbf{k}} = \begin{pmatrix} i\varepsilon_n - \xi(\mathbf{k}) & -\Delta \\ -\Delta & i\varepsilon_n + \xi(\mathbf{k}) \end{pmatrix}.$$

To calculate the averaged Green's functions it is convenient to use a diagram technique.[6] The important difference between our problem and the case of potential scattering by impurities is that the simplest diagram with one cross no longer reduces to an insignificant increment to the ground-state energy. Because of the off-diagonal disorder, the contribution $\overline{V}_{\mathbf{k}\mathbf{k}'} = xb(\mathbf{k}, \mathbf{k})$ of such a diagram leads to a change of the electron spectrum; we note that this contribution is of first order in the fluctuation δb. The importance of effects of first order in the impurity potential in the problem of the metal-to-dielectric transition was noted earlier by Borisyuk.[7] We confine ourselves to the case $|\delta b| \sim \langle |\tilde{u}(\theta)|^2 \rangle N_0$, where N_0 is the state density of states on the Fermi surface, and the angle brackets denote averaging over the angles; therefore $|\delta b| \ll u \equiv (\langle |\tilde{u}(\theta)|^2 \rangle)^{1/2}$ and in all diagrams of second order in $V_{\mathbf{k}\mathbf{k}'}$ the terms with δb are disregarded.

Besides the usual impurity mass operator[6] shown in Fig. 1a, in the case of off-diagonal scattering it is necessary to consider a mass operator of type 1b. The diagonal part of the mass operator is calculated in the same form as in Refs. 3 and 4, and is equal to

$$\hat{\Sigma}_d(\mathbf{k}, i\varepsilon_n) = \begin{pmatrix} \Sigma_1(\mathbf{k}, i\varepsilon_n) & \Sigma_2(\mathbf{k}, i\varepsilon_n) \\ \Sigma_2(\mathbf{k}, i\varepsilon_n) & \Sigma_1(\mathbf{k}-\mathbf{Q}, i\varepsilon_n) \end{pmatrix},$$

$$\Sigma_1(\mathbf{k}, i\varepsilon_n) = \frac{1}{N} \sum_{\mathbf{p}} |\tilde{u}(\mathbf{k}-\mathbf{p})|^2 G(\mathbf{p}, i\varepsilon_n),$$

$$\Sigma_2(\mathbf{k}, i\varepsilon_n) = \frac{1}{N} \sum_{\mathbf{p}} |\tilde{u}(\mathbf{k}-\mathbf{p})|^2 F(\mathbf{p}, i\varepsilon_n).$$

The off-diagonal contribution to the mass operator is

$$\hat{\Sigma}_{od}(\mathbf{k}) = x \begin{pmatrix} b(\mathbf{k}, \mathbf{k}) & b(\mathbf{k}, \mathbf{k}-\mathbf{Q}) \\ b(\mathbf{k}-\mathbf{Q}, \mathbf{k}) & b(\mathbf{k}-\mathbf{Q}, \mathbf{k}-\mathbf{Q}) \end{pmatrix} = x \begin{pmatrix} b(\mathbf{k}, \mathbf{k}) & 0 \\ 0 & -b(\mathbf{k}, \mathbf{k}) \end{pmatrix}.$$

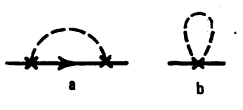

FIG. 1. Contribution made to the mass operator by the diagonal (a) and off-diagonal (b) scattering. The cross denotes the vertex matrix $\hat{V}$, and the line the Green's function $\hat{G}$.

To obtain the last equality we used formulas (5) and (1). The vanishing of the elements $\hat{\Sigma}_{od}$ that are off-diagonal in the sublattice indices is an important factor of the theory, for otherwise the off-diagonal scattering would lead to formation of a gap even in the absence of electron-phonon interaction, and the transition temperature would tend formally to infinity. The averaged Green's function is thus

$$\hat{G}^{-1}(\mathbf{k}, i\varepsilon_n) = \hat{G}_0^{-1}(\mathbf{k}, i\varepsilon_n) - \hat{\Sigma}_d(\mathbf{k}, i\varepsilon_n) - \hat{\Sigma}_{od}(\mathbf{k}) = \begin{pmatrix} i\tilde{\varepsilon}_n(\mathbf{k}) - \tilde{\xi}(\mathbf{k}) & -\tilde{\Delta}_n(\mathbf{k}) \\ -\tilde{\Delta}_n(\mathbf{k}) & i\tilde{\varepsilon}_n(\mathbf{k}) + \tilde{\xi}(\mathbf{k}) \end{pmatrix} \tag{6}$$

Here

$$i\tilde{\varepsilon}_n(\mathbf{k}) = i\varepsilon_n - \Sigma_1(\mathbf{k}, i\varepsilon_n), \quad \tilde{\Delta}_n(\mathbf{k}) = \Delta + \Sigma_2(\mathbf{k}, i\varepsilon_n),$$
$$\tilde{\xi}(\mathbf{k}) = \xi(\mathbf{k}) + xb(\mathbf{k}, \mathbf{k}) = \xi(\mathbf{k})(1 + x\delta b/b). \tag{7}$$

Assuming that in the vicinity of the Fermi energy the dependence of $\tilde{u}(\mathbf{k}-\mathbf{p})$ on $|\mathbf{k}-\mathbf{p}|$ is weak,[6] we find that Σ_1 and Σ_2 depend only on the frequencies

$$\Sigma_1(i\varepsilon_n) = \frac{-i\tilde{\varepsilon}_n}{2\tau(\tilde{\varepsilon}_n^2 + \tilde{\Delta}_n^2)^{1/2}}, \quad \Sigma_2(i\varepsilon_n) = \frac{-\tilde{\Delta}_n}{2\tau(\tilde{\varepsilon}_n^2 + \tilde{\Delta}_n^2)^{1/2}},$$
$$\frac{1}{\tau} = \frac{x}{2} N_0 \int |\tilde{u}(\theta)|^2 d\Omega. \tag{8}$$

Using (7) and (8), we find that

$$\tilde{\varepsilon}_n = \varepsilon_n + u_n/2\tau(1+u_n^2)^{1/2}, \quad \tilde{\Delta}_n = \Delta - 1/2\tau(1+u_n^2)^{1/2},$$

where $u_n = \tilde{\varepsilon}_n/\tilde{\Delta}_n$ is determined by the equation

$$\frac{\varepsilon_n}{\Delta_n} = u_n \left(1 - \frac{1}{\tau\Delta(1+u_n^2)^{1/2}}\right).$$

Thus, our analysis differs from the case of magnetic impurities in superconductors[4] and charged impurities in excitonic dielectrics[3] only in a renormalization of the $\tilde{\xi}(\mathbf{k})$ spectrum; therefore, without dwelling on the details of the solution of the self-consistency equation

$$\Delta = -\frac{g^2(\mathbf{Q})}{\omega_{\mathbf{Q}}} T \sum_{\mathbf{k}n} F(\mathbf{k}, i\varepsilon_n), \tag{9}$$

we present directly the calculation results.

3. DEPENDENCE OF THE GAP AND OF THE TRANSITION TEMPERATURE ON THE IMPURITY CONCENTRATION

Transition from summation over the momentum in (9) to integration over the energies gives rise to a dimensionless interaction constant

$$\lambda = \frac{g^2(Q)}{\omega_Q} N_0 \left(1 - x\frac{\delta b}{b}\right),$$

that depends on the impurity concentration. Inasmuch as substitution changes both the mass of the ion and the electron-ion potential, it follows that λ depends on x not only explicitly because of the factor $1 - x\delta b/b$, but possibly also in a more complicated manner. We put $\lambda_0 = \lambda(x=0)$.

At $T=0$ we get $\Delta_0 = \Delta(T=0)$ from (9):

$$\ln\left[\left(\frac{2W}{\Delta_{00}}\right)^{\lambda_0/\lambda} \frac{\Delta_0(1 - x\delta b/b)}{2W}\right] = \begin{cases} -\pi/4\tau\Delta_{00}, & 1/\tau\Delta_0 < 1 \\ \ln\dfrac{\tau\Delta_0}{1+(1-(\tau\Delta_0)^2)^{1/2}} + \dfrac{1}{2}(1-(\tau\Delta_0)^2)^{1/2} - \dfrac{1}{2\tau\Delta_0}\arcsin\tau\Delta_0, & \dfrac{1}{\tau\Delta_0} > 1 \end{cases} \tag{10}$$

Here $W = |b|z$ is the half-width of the band, and

$$\Delta_{00} = 2W \exp(-1/\lambda_0)$$

is the gap at $T = 0$ for the pure substance.

In the limit $x \ll 1$ it follows from (10) that

$$\Delta_0 = \Delta_{00} - \frac{\pi}{4\tau} + x\Delta_{00}\left(\frac{\delta b}{b} + \frac{1}{\lambda_0^2}\frac{d\lambda}{dx}\right), \quad (11)$$

i.e., increments linear in x are added to the ordinary relations.[3,4] The critical value x_{cr} at which $\Delta_0 = 0$ is given by

$$\tau_{cr}\Delta_{00} = 2\left[1 - x\left(\frac{\delta b}{b} + \frac{1}{\lambda_0^2}\frac{d\lambda}{dx}\right)\right]. \quad (12)$$

Solving (9) in the region $T \lesssim T_c$, we get

$$\ln\left\{T_c\left[1 - x\left(\frac{\delta b}{b} + \frac{1}{\lambda_0^2}\frac{d\lambda}{dx}\right)\right] / T_{c0}\right\} = \psi\left(\frac{1}{2} + \frac{1}{2\pi\tau T_c}\right) - \psi\left(\frac{1}{2}\right), \quad (13)$$

where

$$T_{c0} = \frac{2\gamma}{\pi} W \exp\left(-\frac{1}{\lambda_0}\right)$$

is the temperature of the transition in pure matter, and $\psi(x)$ is the derivative of the logarithm of the Γ function. At $x \ll 1$ we have

$$T_c = T_{c0} - \frac{\pi}{4\tau} + xT_{c0}\left(\frac{\delta b}{b} + \frac{1}{\lambda_0^2}\frac{d\lambda}{dx}\right). \quad (14)$$

From the condition $T_c = 0$ we get

$$\tau_{cr}T_{c0} = \frac{2\gamma}{\pi}\left[1 - x\left(\frac{\delta b}{b} + \frac{1}{\lambda_0^2}\frac{d\lambda}{dx}\right)\right],$$

which coincides with (12).

4. DISCUSSION OF RESULTS

The proposed theory is applicable to transition-metal compounds in which a metal-to-dielectric transition with lattice distortion is observed. As a rule, these substances make up large groups of isostructural compounds, for example the dioxides MeO_2 with rutile structure, the sesquioxides Me_2O_3 with corundum structure, the monosulfides MeS, and others.[8] Within each group it is possible to synthesize a large number of disordered alloys, and if the $3d$ ion is replaced by a $4d$ or $5d$ ion with larger dimension, then the hopping integrals increase, i.e. $\delta b > 0$. On the other hand if a $4d$ ion is replaced by a $3d$ ion, then $\delta b < 0$. In the former case Δ and T_c have a tendency to increase. The physical reason is the increased width of the band when the $3d$ ion is replaced by $4d$.

It is of interest to investigate the system $V_{1-x}Nb_xO_2$ at $x \ll 1$ ($\delta b > 0$) and at $x \lesssim 1$, $1 - x \ll 1$ ($\delta b < 0$). In the former case the decrease of T_c would be very slow (or nonexistent), in the latter T_c would drop steeply. We know of experimental results for two other alloys: $V_{1-x}Mo_xO_2$,[1)] where a noticeable decrease of T_c occurs only at $x \sim 0.1$, and $Nb_{1-x}Ti_xO_2$ (Ref. 9), where $T_{c0} = 1090$ K, and at $x = 0.017$ the transition temperature drops to $T_c = 300$ K, so that in the second case the drop is very rapid. Such a substitution gives rise to excess carriers—electrons in the case of Mo and holes in the case of Ti. According to the band model of dioxides,[10] the extra Mo electrons fill the e_g band and not change the filling of the quasi-one-dimensional a_1 band, i.e., they do not violate the condition $\mu = \varepsilon$, and the theory can be used without any changes. The holes from Ti, however, decrease the occupation of the a_1 band and we get $\delta\mu = \varepsilon - \mu > 0$, which also leads to a decrease of T_c.

When the $3d$ ion is replaced by a $5d$ ion, the fluctuations of $\delta b/b$ should increase, one should expect an even slower decrease of T_c than in $V_{1-x}Mo_xO_2$, or even an increase of T_c. In the considered compounds $T_{c0} \sim 10^2 - 10^3$ K, therefore the critical value of the concentration [(formula (12)] will not be reached in the region $x \ll 1$ where the theory is valid; the structurally distorted phase can apparently exist in the entire concentration interval. This conclusion is valid for the compound $V_{1-x}Mo_xO_2$ at least up to $x = 0.2$ (Ref. 11).

Assuming, following Zittartz,[3] that $\tau = 10^{-11}$ sec at $x = 0.01$, we can estimate from the data of Horlin *et al.*[11] the parameter δb (neglecting the dependence of λ on x); this yields $\delta b N_0 = 1/40$. On the other hand $\tau^{-1} = x\mu^2 N_0$, whence $\mu N_0 \sim 0.1$ at a band width $2W \sim N_0^{-1} \sim 1$ eV. These estimates show that the condition $|\delta b| \ll \mu$ used in Sec. 2 can be satisfied at reasonable values of the parameters.

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