Combined potential and spin impurity scattering in cuprates

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We present a theory of combined nonmagnetic and magnetic impurity scattering in anisotropic superconductors accounting for the momentum-dependent impurity potential. Applying the model to the d-wave superconducting state, we obtain a quantitative agreement with the initial suppression of the critical temperature due to Zn and Ni substitutions as well as electron irradiation defects in the cuprates. We suggest that the unequal pair-breaking effect of Zn and Ni may be related to a different nature of the magnetic moments induced by these impurities.

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Impurities offer a useful experimental probe of the fundamental properties of high-temperature superconductors. Controlled substitutions of $3d$ transition metals (Zn, Ni) provide indirect information on the nature of the pairing mechanism since they affect the physical properties of both the superconducting and normal state. The nominally nonmagnetic Zn ($3d^{10}$, $S=0$) as well as magnetic Ni ($3d^8$, $S=1$) atoms reside in the magnetically active in-plane Cu ($3d^9$, $S=1/2$) sites. Macroscopic susceptibility and NMR measurements[1–5] of $YBa_2Cu_3O_{7-\delta}$ (Y-123) superconductor indicate the impurity-induced magnetic moments of $0.86\mu_B$ for Zn, and $1.9\mu_B$ for Ni in the underdoped compound, which decrease with hole doping to $0.36\mu_B$/Zn and $1.6\mu_B$/Ni in the optimally doped system. In $La_{2-x}Sr_xCuO_4$ (La-214) the same impurity substitutions lead to magnetic properties corresponding to local magnetic moments of $1.0\mu_B$/Zn and $0.6\mu_B$/Ni.[6] Magnetism generated by Zn and Ni atoms is, however, different in nature. Whereas, the Zn-induced moments reside in the vicinity of the nonmagnetic impurity on the neighboring Cu sites,[3–5,7] the Ni substitution yields the magnetic moments which partially screen $S=1$ impurity spin and result in a more localized $S=1/2$ moment.[5,7] In the cuprates, where the strong Coulomb interactions are present, a creation of a magnetic moment around any lattice defect cannot be excluded. Particularly, a removal of the oxygen atom from the CuO_2 plane,[8–11] where the Cu ($3d$) spins and O ($2p$) holes are strongly coupled, should give rise to a local uncompensated magnetic moment, that is it should develop a magnetically active oxygen vacancy defect.

A theoretical analysis of the impurity influence on the d-wave superconducting state based on a standard isotropic (s-wave) scattering approach indicates a significantly stronger suppression of superconductivity than observed in the cuprates.[12] This discrepancy can be reconciled within a simple model of anisotropic impurity scattering[13,14] which takes also the non-s-wave scattering channels into account. Evaluation of the critical temperature[13] and the upper critical field[15,16] dependences on the scattering rate showed their robustness to impurity scattering in the d-wave channel and led to a quantitative agreement with the experimental data. A strong support to these early theoretical predictions is provided by a recent experiment[17] with the electron irradiation created oxygen defects in Y-123 which confirms the extended nature of the scattering centers and the ability of the model to account for both the T_c and H_{c_2} suppression by the disorder. Despite its preliminary success this approach neglects the magnetic scattering processes which however should be present at least for Zn and Ni impurity substitutions. Obviously, the spin-flip scattering leads to an additional destruction of a singlet d-wave superconducting state and, if included in our model, could possibly shift its predictions beyond the experimental range. In this paper, we discuss this issue in more detail by generalizing the effective anisotropic impurity scattering[13,14] to a combined potential and magnetic scattering. We analyze the initial T_c suppression in $YBa_2Cu_3O_{7-\delta}$, $La_{2-x}Sr_xCuO_4$, $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) compounds induced by Zn (Refs. 18–24) and Ni (Refs. 22,25) substitution as well as by the in-plane oxygen vacancy defect created by the electron irradiation.[8–11] Discussing the T_c alone cannot give a final test of the impurity scattering model. Its applicability can be ultimately verified by a thorough analysis of the thermodynamic and transport properties as has been described for isotropic scattering in Refs. 26–28. Such a comprehensive approach is beyond the scope of this Brief Report in which we focus on a possible role of the extended magnetic scattering in the suppression of the critical temperature. An elaborated study of the presented model will be given in a separate paper.

We consider randomly distributed impurities at low concentration interacting with conduction electrons through a potential $u(\mathbf{k},\mathbf{k}')=v(\mathbf{k},\mathbf{k}')+J(\mathbf{k},\mathbf{k}')\overline{\mathbf{S}\sigma}$, where $\overline{\mathbf{S}}$ is a classical spin representing the impurity and $\overline{\boldsymbol{\sigma}}$ is the electron spin density. The anisotropic superconducting state is determined by the order parameter $\Delta(\mathbf{k})=\Delta e(\mathbf{k})$, where $e(\mathbf{k})$ is a momentum-dependent function. We normalize $e(\mathbf{k})$ in this way that $\langle e^2\rangle=1$, where $\langle\cdots\rangle=\int_{FS}dS_k n(\mathbf{k})(\cdots)$ denotes the average value over the Fermi surface (FS) momenta, and $n(\mathbf{k})$ is the angle resolved FS density of states normalized to unity, i.e., $\int_{FS}dS_k n(\mathbf{k})=1$. For the $(d_{x^2-y^2}+s)$-wave state $e(\mathbf{k})=(\cos 2\phi+s)/\langle(\cos 2\phi+s)^2\rangle^{1/2}$ in a polar angle notation where the $d_{x^2-y^2}$-wave state corresponds to $s=0$. Taking the electron-impurity scattering within the Born approximation and neglecting the impurity-impurity interaction,[29] the diagonal and off-diagonal corrections to the Green's function averaged over the impurity positions and spin directions read[29,30]

$$\tilde{\omega}(\mathbf{k})=\omega+\pi n_i N_0\int_{\mathrm{FS}} dS_{\mathbf{k}'} n(\mathbf{k}')\tilde{\omega}(\mathbf{k}') \times\frac{[v^2(\mathbf{k},\mathbf{k}')+J^2(\mathbf{k},\mathbf{k}')S(S+1)]}{[\tilde{\omega}^2(\mathbf{k}')+\tilde{\Delta}^2(\mathbf{k}')]^{1/2}}, \qquad (1)$$

$$\tilde{\Delta}(\mathbf{k})=\Delta(\mathbf{k})+\pi n_i N_0\int_{\mathrm{FS}} dS_{\mathbf{k}'} n(\mathbf{k}')\tilde{\Delta}(\mathbf{k}') \times\frac{[v^2(\mathbf{k},\mathbf{k}')-J^2(\mathbf{k},\mathbf{k}')S(S+1)]}{[\tilde{\omega}^2(\mathbf{k}')+\tilde{\Delta}^2(\mathbf{k}')]^{1/2}}, \qquad (2)$$

where $\omega=\pi T(2n+1)$ is the Matsubara frequency (T is temperature and n is an integer number), N_0 is the single-spin density of states at the Fermi level, n_i is the impurity (defect) concentration, and all the wave vectors are restricted to the Fermi surface. We generalize our earlier model[13,14] to include magnetic scattering and assume that the momentum-dependent potential terms in Eqs. (1),(2) are separable and given by

$$v^2(\mathbf{k},\mathbf{k}')=v_0^2+v_1^2 f(\mathbf{k})f(\mathbf{k}'), \qquad (3)$$

$$J^2(\mathbf{k},\mathbf{k}')=J_0^2+J_1^2 g(\mathbf{k})g(\mathbf{k}'), \qquad (4)$$

where v_0 (v_1), J_0 (J_1) are isotropic (anisotropic) scattering amplitudes and $f(\mathbf{k})$, $g(\mathbf{k})$ are the momentum-dependent anisotropy functions in the nonmagnetic and magnetic scattering channel, respectively.

We assume that the overall scattering rate is determined by the isotropic components and to have it well defined impose the constraints

$$v_1^2 \leqslant v_0^2, \quad \langle f\rangle=0, \quad \langle f^2\rangle=1, \qquad (5)$$

$$J_1^2 \leqslant J_0^2, \quad \langle g\rangle=0, \quad \langle g^2\rangle=1. \qquad (6)$$

Note, that such a choice of the anisotropy does not change the normal state properties as the diagonal part of the self-energy in the limit of $\Delta\to 0$ is given by $\tilde{\omega}(\mathbf{k})_{\Delta=0}=\omega+\pi n_i N_0[v_0^2+J_0^2 S(S+1)]\mathrm{sgn}(\omega)$ and depends only on the isotropic scattering rates.

Inclusion of a momentum-dependent model impurity potential in the magnetic channel is motivated by the existence of extended magnetic moments associated with the impurity,[1–7] and the anisotropy of the nonmagnetic scattering channel simulates the anisotropy of the crystal lattice. For Ni impurity which leads to a screened localized magnetic moment[5,7] we expect $J_1/J_0 \ll 1$, while for broadly distributed over Cu sites Zn-induced magnetic moment[3–5,7] both isotropic and anisotropic scattering channels should be comparable in magnitude, i.e., $J_1/J_0 \sim 1$. Contrary to the NMR measurements[2–5] muon spin rotation (μSR) experiments[31] report no evidence for additional Zn-induced moments in underdoped Y-123. Yet, one cannot exclude the possibility that the impurity-generated magnetic moments exist but are weakly coupled to the spin system of the CuO_2 planes. Therefore, the following assumption about the relative amount of magnetic scattering seems reasonable: $J_1/v_1 < 1$, $J_0/v_0 < 1$.

Taking a separable pair potential $V(\mathbf{k},\mathbf{k}')=-V_0 e(\mathbf{k})e(\mathbf{k}')$, $V_0>0$, and following a standard procedure[13,30] we obtain the critical temperature T_c equation

$$\ln\frac{T_c}{T_{c_0}}=(1-\langle e\rangle^2)\left[\psi\left(\frac{1}{2}\right)-\psi\left(\frac{1}{2}+\frac{\Gamma_0+G_0}{2\pi T_c}\right)\right] +\langle e\rangle^2\left[\psi\left(\frac{1}{2}\right)-\psi\left(\frac{1}{2}+\frac{2G_0}{2\pi T_c}\right)\right]+S_1+S_2, \qquad (7)$$

where T_{c_0} is the critical temperature in the absence of impurities, $\psi(z)$ is the digamma function,

$$S_1=2\pi T_c\sum_{\omega>0}[\langle ef\rangle/(\omega+\Gamma_0+G_0)] \times\frac{\Gamma_1\langle ef\rangle(\omega+\Gamma_0+G_0+G_1)-\Gamma_1 G_1\langle eg\rangle\langle fg\rangle}{(\omega+\Gamma_0+G_0+G_1)(\omega+\Gamma_0+G_0-\Gamma_1)+G_1\Gamma_1\langle fg\rangle^2},$$

$$S_2=-2\pi T_c\sum_{\omega>0}[\langle eg\rangle/(\omega+\Gamma_0+G_0)] \times\frac{G_1\langle eg\rangle(\omega+\Gamma_0+G_0-\Gamma_1)+\Gamma_1 G_1\langle ef\rangle\langle fg\rangle}{(\omega+\Gamma_0+G_0+G_1)(\omega+\Gamma_0+G_0-\Gamma_1)+G_1\Gamma_1\langle fg\rangle^2},$$

and $\Gamma_0=\pi n_i N_0 v_0^2$, $\Gamma_1=\pi n_i N_0 v_1^2$, $G_0=\pi n_i N_0 J_0^2 S(S+1)$, and $G_1=\pi n_i N_0 J_1^2 S(S+1)$ are the intrinsic scattering rates. Given strongly coupled spin and charge dynamics in the copper-oxygen planes the impurity potential encountered by the electron in the spin scattering channel should follow the one in the nonmagnetic channel, that is $g(\mathbf{k})=\pm f(\mathbf{k})$, which simplifies Eq. (7) to

$$\ln\frac{T_c}{T_{c_0}}=(1-\langle e\rangle^2-\langle ef\rangle^2)\left[\psi\left(\frac{1}{2}\right)-\psi\left(\frac{1}{2}+\frac{\Gamma_0+G_0}{2\pi T_c}\right)\right] +\langle ef\rangle^2\left[\psi\left(\frac{1}{2}\right)-\psi\left(\frac{1}{2}+\frac{\Gamma_0+G_0+G_1-\Gamma_1}{2\pi T_c}\right)\right] +\langle e\rangle^2\left[\psi\left(\frac{1}{2}\right)-\psi\left(\frac{1}{2}+\frac{2G_0}{2\pi T_c}\right)\right]. \qquad (8)$$

Comparing the above equation with the one for nonmagnetic scattering [Eq. (24) of Ref. 13] we notice that there is only one additional term in Eq. (8) reflecting an extra T_c suppression due to the spin-flip scattering determined by the exchange rate $2G_0$. The remaining terms of Eq. (8) correspond to the ones obtained for potential scattering[13] with some new effective scattering rates being combinations of the scattering rates in the magnetic and nonmagnetic channels $\overline{\Gamma}_0=\Gamma_0+G_0$, $\overline{\Gamma}_1=\Gamma_1-G_1$. We note, that the anisotropy of the impurity potential has no effect on the T_c when $\overline{\Gamma}_1=0$, i.e., when $\Gamma_1=G_1$, which should not be the case of the cuprates, where most probably the exchange interaction between the impurity spin and the Cu-O spin system is small $G_1/\Gamma_1<1$. The role of anisotropy in the impurity potential is concurrently expressed by the dimensionless parameter $\langle ef\rangle^2$ $(0\leqslant\langle ef\rangle^2\leqslant 1)$ representing the interplay between the pair potential and the anisotropic part of the impurity potential.

TABLE I. ϕ_A anisotropy factor of the impurity potential reproducing the T_c suppression in the Y-123 compound within the *d*-wave superconductivity scenario, i.e., $\chi=1$. (1.1 eV$\leqslant\omega_{\rm pl}\leqslant$1.4 eV; ov.=overdoped, op.=optimally doped, un.=underdoped.)

Defect	Sample	$(dT_c/d\rho_0)_{\rm exp}$ [K/$\mu\Omega$ cm]	Anisotropy factor ϕ_A
	(ov.) single crystal (Ref. 18)	-0.674	$0.093\leqslant\phi_A\leqslant0.440$
Zn	(ov.) single crystal (Refs. 19,22)	-0.57	$0.233\leqslant\phi_A\leqslant0.526$
impurity	(op.) thin film (Ref. 23)	-0.241	$0.676\leqslant\phi_A\leqslant0.780$
	(op.) thin film (Ref. 24)	-0.520	$0.300\leqslant\phi_A\leqslant0.568$
Ni	(ov.) ceramic sample (Ref. 22)	-0.333	$0.552\leqslant\phi_A\leqslant0.723$
impurity	(ov.) film (Ref. 25)	$-0.063--0.044$	$0.915\leqslant\phi_A\leqslant0.963$
O	(ov.) single crystal (Ref. 9)	-0.30 ± 0.04	$0.542\leqslant\phi_A\leqslant0.784$
vacany	(ov.) film (Ref. 8)	-0.187	$0.748\leqslant\phi_A\leqslant0.845$

Again, for $\langle ef\rangle^2=0$ we obtain a regular T_c suppression due to the isotropic combined nonmagnetic and magnetic impurity scattering[32] which in the case of a conventional *s*-wave superconductor reduces to the well known relationship.[29,30]

In the quantitative analysis of the experimental data we consider the T_c reduction in the limit of low impurity concentration $n_i\rightarrow0$ which is given by the initial slope

$$dT_c/d\overline{\Gamma}_0=-(\pi/4)[\chi+(1-\chi)\phi_M-\phi_A], \qquad (9)$$

where for the sake of brevity we have introduced $\chi=1-\langle e\rangle^2$, $\phi_A=\langle ef\rangle^2\overline{\Gamma}_1/\overline{\Gamma}_0$ (anisotropy factor), and $\phi_M=2G_0/\overline{\Gamma}_0$ (magnetic factor). The isotropic part of the impurity potential $\overline{\Gamma}_0$ determines the residual resistivity at the zero frequency[13] $\rho_0\simeq10.18\times10^{-2}$ $(8\pi^2/\omega_{\rm pl}^2)T_{c_0}(\overline{\Gamma}_0/2\pi T_{c_0})\mu\Omega$ cm, where $\omega_{\rm pl}$ is the in-plane plasma frequency in eV. We fix $\omega_{\rm pl}$ within the range[12] 1.1 eV$\leqslant\omega_{\rm pl}\leqslant$1.4 eV for Y-123, and estimate it from the zero temperature penetration depth measurements[33–35] as 0.84 and 0.9 eV for La-214 and Bi-2212, respectively. There is still no complete experimental agreement on the impurity-induced resistivity in the cuprates.[18,19,22,24,25] However, some results[18,22] suggest that Zn and Ni lead to a comparable effect in the normal state, which would mean that $\overline{\Gamma}_0(\mathrm{Zn})\sim\overline{\Gamma}_0(\mathrm{Ni})$. The residual resistivity ρ_0 is used to express the initial T_c suppression in the following form convenient for the discussion of the experimental data:

$$dT_c/d\rho_0\simeq-0.614\ \ \omega_{\rm pl}^2[\chi+(1-\chi)\phi_M-\phi_A]\ \mathrm{K}/\mu\Omega\ \mathrm{cm}. \qquad (10)$$

We find that in the *d*-wave state ($\chi=1$) the initial T_c suppression [Eq. (10)] is determined only by the dimensionless anisotropy factor ϕ_A. The influence of the spin-flip scattering is present through the renormalized scattering rates in the isotropic and anisotropic scattering channels $\overline{\Gamma}_0$ and $\overline{\Gamma}_1$. However, the magnetic factor ϕ_M has no effect on the critical temperature and we may say that the *d*-wave system becomes insensitive to magnetic scattering as the solutions of Eq. (8) correspond to those for nonmagnetic scattering[13] with renormalized parameters. In Tables I–III we give the anisotropy factors ϕ_A needed to account for the experimental data of Zn, Ni, and O vacancy-induced T_c suppression in Y-123, La-214, and Bi-2212 compounds. Worth noting are generally significantly lower values of ϕ_A for Zn substitution than for Ni. This feature can be attributed to a different nature of the magnetic moments associated with these impurities. While the Zn-induced moment is broadly distributed over neighboring Cu sites ($G_1/G_0\sim1$), the one created by Ni is screened on a larger distance and is more localized ($G_1/G_0\ll1$). Therefore, for comparable amounts of isotropic scattering by these impurities, $\overline{\Gamma}_0=\Gamma_0+G_0$, the anisotropy factor $\phi_A\sim(\Gamma_1-G_1)/(\Gamma_0+G_0)$ of Ni scattering potential, $\sim\Gamma_1/(\Gamma_0+G_0)$, should exceed the one of Zn impurity $\sim(\Gamma_1-G_0)/(\Gamma_0+G_0)$. Given large ϕ_A values for the oxygen vacancy we may also infer that the magnetic moments formed around this defect are rather localized or the anisotropic scattering takes place in the *d*-wave channel, i.e., $\langle ef\rangle^2\sim1$. Still, to decide definitely about the role of spin scattering in high-T_c materials with simple defects a quantitative estimate of the ratios of the magnetic to nonmagnetic scattering rates G_0/Γ_0 and G_1/Γ_1 is required. Of some help here may be a

TABLE II. ϕ_A anisotropy factor of the impurity potential reproducing the T_c suppression in the La-214 compound within the *d*-wave superconductivity scenario, i.e., $\chi=1$. ($\omega_{\rm pl}=0.84$ eV; notation as in Table I.)

Defect	Sample	$(dT_c/d\rho_0)_{\rm exp}$ [K/$\mu\Omega$ cm]	Anisotropy factor ϕ_A
Zn	(ov.) single crystal (Ref. 19)	-0.37	0.146
impurity	(un.) film (Refs. 20,21)	-0.233	0.462
O vacancy	(un.) film (Ref. 10)	-0.127	0.707

TABLE III. ϕ_A anisotropy factor of the impurity potential reproducing the T_c suppression in the Bi-2212 compound within the *d*-wave superconductivity scenario, i.e., $\chi=1$. ($\omega_{\rm pl}=0.9$ eV; notation as in Table I.)

Defect	Sample	$(dT_c/d\rho_0)_{\rm exp}$ [K/$\mu\Omega$ cm]	Anisotropy factor ϕ_A
O vacancy	(ov.) single crystal (Ref. 11)	-0.28	0.437

generalization of the H_{c_2} critical field analysis to magnetic impurity scattering. So far, an application of the nonmagnetic scattering expression for the reduced H_{c_2} slope[15,16] in the interpretation of the critical field of the electron irradiated Y-123 has confirmed the anisotropy parameters of the model determined from T_c suppression in this compound.[17]

The orthorhombic distortion of the crystal lattice in the Y-123 superconductor leads to the $(d+s)$-wave superconductivity in this compound.[36] The photoemission, tunneling, and thermodynamic measurements put an upper bound of $\sim$10% on the relative size of the *s*-wave component in the $(d+s)$-wave superconducting state.[36] If we define a per cent fraction $\Delta_{s\%}$ of the *s*-wave component as a ratio of a minimal gap to its maximum value, then the condition $\Delta_{s\%}=10\%$ is equivalent to $\chi=0.976$ ($\langle e\rangle^2=0.0241$). It is a matter of a straightforward calculation to show that ϕ_A for a given superconducting state is related to the one of the *d*-wave state ϕ_A^0 through $\phi_A=\phi_A^0-(1-\chi)(1-\phi_M)$. In what follows, for $\chi=0.976$ we get merely a slight change of the anisotropy factor $\phi_A=\phi_A^0-0.024(1-\phi_M)$ which for different amounts of the magnetic scattering rate reads $\phi_A=\phi_A^0-0.024$ for $\phi_M=0$ ($G_0=0$); $\phi_A=\phi_A^0-0.008$ for $\phi_M=0.666$ ($G_0/\Gamma_0=0.5$); and $\phi_A=\phi_A^0$ for $\phi_M=1$ ($G_0/\Gamma_0=1$). Our approach reduces to the isotropic impurity scattering approximation[32] in the limit of $\phi_A=0$. Therefore, given the nonzero ϕ_A values in Tables I–III and taking the above corrections for the $(d+s)$-wave state into account, we note that the experimental data cannot be explained by applying the isotropic impurity scattering approach to the $(d+s)$-wave superconducting state with an appropriate (for the cuprates) amount of the *s*-wave component.

Concluding, we find that even in the presence of the spin-flip processes the scenario of anisotropic impurity scattering accounts quantitatively for the experimentally observed T_c suppression due to Zn, Ni, and O vacancy scattering. Moreover, a different partition of the magnetic scattering rates into isotropic and anisotropic scattering channels can be used in the interpretation of the weak Ni-induced and relatively strong Zn-induced pair breaking. Our result also shows that the isotropic impurity scattering approximation does not explain quantitatively the initial suppression of the critical temperature in the cuprates. Finally, we note that a significant sample dependence of the discussed data with the discrepancy even as high as 50% calls for further more controlled measurements of the impurity-induced T_c suppression in the high-temperature superconductors.

Also, please note that except for the Ni effect in overdopd Y-123 films[25] the anisotropy factor of the *d*-wave state (Tables I–III) can be chosen lower than a cylindrical Fermi surface average of the absolute value of the order parameter $\langle|e|\rangle\simeq0.81$.

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