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Influence of thermal fluctuations on single-vortex pinning in Rb_3C_{60} fullerene superconductors

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From the experimentally obtained $H(T)$ dependence of the transition boundary between short-range and long-range vortex order for Rb_3C_{60} superconductors, the temperature dependences of the effective potential barrier for pinning of a single vortex on a pinning center and the effective pinning force are calculated. It is shown that the effective barrier falls almost linearly with the increase of temperature and that the harmonic thermal fluctuations strongly decrease the effective pinning force as $T^{3/2}$.

A large region of Shubnikov mixed state is characteristic of alkali-metal-doped fullerenes which are strong type-II superconductors. For applied fields between the first (H_{c1}) and the second (H_{c2}) critical fields, quantized flux tubes are formed inside the sample. In the absence of disorder they form a regular lattice of parallel flux lines or vortices, the vortex lattice (VLL). Disorder in the crystal structure of a superconductor, such as defects, twinning boundaries, inhomogeneities, etc., leads to pinning of vortex lines, and the crystalline long-range order of the vortex-line lattice as was shown in Refs. 1 and 2 is unstable in the presence of randomly distributed pinning centers. The magnetic properties of a superconductor reflect all these processes. The magnetic behavior of fullerene superconductors shows several unusual properties, such as very rapid fall of the critical-current density (J_c) with temperature[3] and magnetic field.[4] Thermal fluctuations play an important role in these phenomena. The total combination of factors (a) high critical transition temperature ($T_c \simeq 20$–40 K), (b) short coherence length ($\xi \simeq 30$ Å), and (c) large penetration depth ($\lambda \simeq 2500$ Å) leads to an enhancement of the effect of thermal fluctuations. In Ref. 5 it was shown that in extreme type-II superconductors phononlike harmonic thermal fluctuations of vortex lines reduce the effective pinning strength and hence strongly reduce the value of J_c. The effects of fluctuations are remarkable not only in strong magnetic fields but even close to H_{c1}.[6] There are three energies most important in the consideration of pinning of the vortex lines: pinning energy, repulsive intervortex interaction energy, and an entropic contribution.

It is evident that due to a random distribution of pinning centers their positions do not coincide with the positions of the vortices in the VLL. Therefore pinned vortices are displaced with respect to the vortex lattice sites. That leads to destruction of long-range order; instead, a short-range order (or vortex glass) exists.

A pinned vortex is in a potential well with a pinning barrier $U_{\rm pin}$. The effective barrier height $U_{\rm eff} = U_{\rm pin} - kT$ is dependent on competition between thermal fluctuations and the pinning force. When the repulsive energy becomes equal to $U_{\rm eff}$ the pinning force is overcome and the vortex takes its place in the VLL. The higher the temperature the smaller the effective potential barrier; hence a smaller repulsive energy is necessary to unpin the vortex. A vortex line at a finite temperature can, with some probability, overcome the potential barrier that is created by a pinning center. If the gradient of the magnetic pressure is not zero ($\delta p/\delta x \neq 0$) then vortex lines move to regions with smaller values of the magnetic induction. The interaction of a vortex with a cylindrical volume of diameter $r > \xi$ was considered in Ref. 7. Because the core of the vortex is in the normal state and the free energy of the normal state is higher than that of the superconducting state, the movement of the vortex from a normal to a superconducting volume leads to an increase of energy, $\Delta W = wV$. Here, $w = H_{cm}^2/8\pi$ is the volumetric energy density, H_{cm} is the critical field of the bulk material, and V is the volume of the vortex. For a unit vortex length $\Delta W = h_{cm}^2 \xi^2/8$.[7] This is the barrier that a vortex has to overcome as it moves from a normal to a superconducting volume. The work of the movement over a distance about ξ is equal to ΔW. Therefore the average force f_p for a volume of diameter r is[7]

$$f_p \simeq H_{cm}^2 \xi r/8 \ , \tag{1}$$

where both H_{cm}^2 and ξ are temperature dependent. Close to the critical temperature the $H_{cm}(T)$ and $\xi(T)$ dependences are known from the Ginzburg-Landau theory and the equation for $f(p)$ is

$$f_p \simeq \frac{\phi_0^2(1-T/T_c)^{3/2}}{64\pi^2\lambda_0^2\xi_0} r \ , \tag{2}$$

where ϕ_0 is the quantum flux and λ is the penetration depth.

At low temperature ($T \ll T_c$) where H_{cm} and ξ are practically temperature independent,[8] the influence of the temperature dependence of r, where r is the linear size of the defect, on the pinning force should be taken into account [see Eq. (1)]. The coefficient of linear expansion is

$$\Lambda = 1/l \times dl/dT \ , \tag{3}$$

where l is a length. On the other hand,[9] $\Lambda = \gamma C_v/3B$, where γ is Grüneisen's parameter and C_v is the specific heat. It is seen that Λ depends on temperature in the same way as C_v. It is known from dielectrics that $C_v \sim T^3$ and $T \to 0$ and C_v is a constant at $T \gg \Theta_p$ (Θ_p is a characterstic temperature). Proceeding from this, one finds $\Lambda = AT^n$, where $3 \geq n \geq 0$. Substituting this in Eq. (3) one gets

$$l = l_0 \times \exp\{\Lambda_0[(T/T_c)^{n+1} - 1]\} \ ,$$

where $\Lambda_0 = AT_0^{n-1}/(n+1)$. Taking into account that the power of the exponent is much less than unity, one obtains

$$l \cong l_0\{1 + \Lambda_0[(T/T_c)^{n+1} - 1]\} \ . \tag{4}$$

The $l(T)$ dependence is changed from $l \sim T^4$ to $l \sim T$ and in the low-temperature region the pinning force increases with temperature just as the linear size of the defect does.

It is necessary to note that all previous equations are approximate and, in the model considered, vortices are acting independently, i.e., there is no correlated motion. The quantitative effect of thermal fluctuations on that process is still controversial from both theoretical and experimental points of view.

In some theoretical and experimental research (see, for instance, Refs. 10–12), the vortex-depinning process and the influence of thermal fluctuations on it were investigated for high-T_c superconductors with strong anisotropy. However, this question for materials with high critical temperature, three-dimensional superconductivity, and a strong pinning potential is unresolved.

In Ref. 4 a transition in a vortex system from short-range to long-range order in the collective vortex-pinning regime was shown. A superconducting Rb_3C_{60} powder sample was investigated by dc magnetization measurements. In that experiment the magnetic-field dependence of the inverse critical-current density was linear. The behavior of $1/J_c(H)$ is in good agreement with collective pinning theory[2,5] which gives the relation between critical-current density and magnetic-field induction (B) as

$$J_c \sim (F_p/V_c)^{1/2} B^{-1} \ , \tag{5}$$

where F_p is the mean square value of the random pinning force, $V_c = R_c^2 L_c$ is the volume of a correlated vortex cluster, and L_c (R_c) is the longitudinal (transverse) size of this cluster. On the transition boundary between short-range and long-range vortex order, at the characteristic value of external magnetic field $\tilde{H}$, kink in the linear dependence of $1/J_c(H)$ was observed. It was shown that the value of the pinning force F_p was a constant in the whole experimental region of external magnetic field and temperature. Hence, following Eq. (5), the changes of slope in the $1/J_c(H)$ dependence lead an increase in volume of the correlated vortex clusters on the transition boundary by a factor of 6. The transverse size of the clusters R_c becomes equal to the size of the superconducting grains in the sample.[4] The transition boundary on the HT diagram is linear (Fig. 1) and can be described by

$$\tilde{H} = 5.6 \times 10^4 \times (1 - T/T_c) \ . \tag{6}$$

At small external magnetic field the vortex system is a short-range ordered structure. With increasing H_{ext} the distance between vortices decreases as $d = (\phi_0/H_{ext})^{1/2}$ and hence the repulsive interaction energy U_{rep} increases. On the transition boundary $\tilde{U}_{rep}$ becomes equal to the effective potential barrier U_{eff} and the pinned vortices overcome the barrier and form a vortex lattice. This means that a transition from a vortex glass to a distorted vortex lattice has occurred. A similar transition from short-range to long-range vortex order in the regime of collective vortex pinning was experimentally observed by Träuble and Essmann.[13]

One can easily estimate the repulsive energy $\tilde{U}_{rep}$ and the repulsive intervortex force $\tilde{f}_{rep}$ on the transition boundary (here and further a tilde shows that the value of a parameter is taken on the transition boundary) and, since at zero temperature $\tilde{U}_{rep}(0) = U_{pin} = U_{eff}(0)$ and $\tilde{f}_{rep} = F_p$, find that the values of U_{pin} and F_p are constant in the whole experimental region of T and H.[4]

For this estimation let us consider the interaction of two parallel flux lines. The force experienced by flux line 1 due to the presence of another, parallel flux line 2 (separated from flux line 1 by a distance d) is given by[14]

$$f_d = (\phi_0^2/8\pi^2\lambda^3) K_1 \left[\frac{d}{\lambda} \right] \ , \tag{7}$$

where K_1 is the modified Hankel function of order one. For $x = d/\lambda \ll 1$ the function $K_1(x)$ may be approximated by

$$K_1(x) \simeq \frac{1}{x} - \frac{(1.23 - 2\ln x)x}{4} \ . \tag{8}$$

For the specimen under consideration $\lambda(0) = 2500$ Å (Ref. 15) and $\tilde{d}(0) = (\phi_0/\tilde{H})^{1/2}$, where $\tilde{H}(0) = 56$ kOe [see Eq. (6)]. Because the repulsive force $\tilde{f}_{rep}$ and pinning force F_p are equal and opposite on the transition bound-

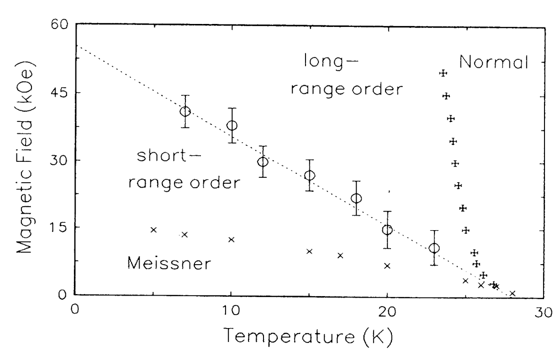

FIG. 1. Phase diagram of Rb_3C_{60} superconductor (Ref. 4). $\times$, values of the lower critical magnetic field (H_{c1}), multiplied by a factor of 100 (Ref. 18); $\circ$, values of $\tilde{H}$; $+$, values of the upper critical magnetic field (H_{c2}) (Ref. 15).

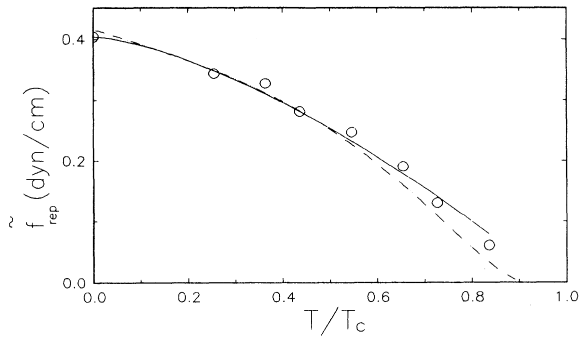

FIG. 2. Temperature dependence of $\tilde{f}_{rep}$; solid line corresponds to Eq. (10) and dotted line to Eq. (11).

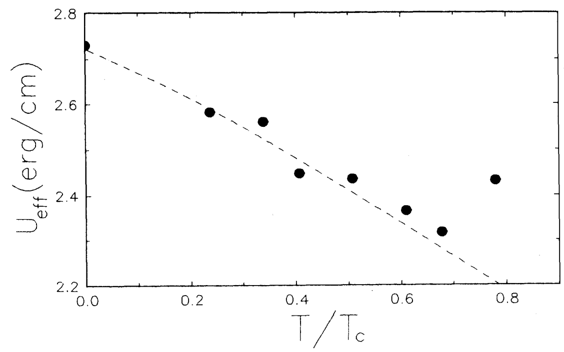

FIG. 3. Temperature dependence of the effective pinning potential barrier; dotted line corresponds to Eq. (14).

ary, using values of $\lambda(0)$ and $\tilde{d}(0)$, we can find the value $F_p = 0.4$ dyn/cm of the pinning force per unit length of a flux line.

From the experimental values of $\tilde{H}$ at different temperatures and $\lambda(T)$ from Ref. 15 using a two-fluid model approximation

$$\lambda(T) = \lambda(0)[1-(T/T_c)^4]^{-1/2} , \tag{9}$$

(which gives good agreement with experimental results),[16,15] one can calculate values of the repulsive intervortex force on the transition boundary which correspond to escape of the pinned vortex from the potential well at different temperatures. The results of this calculation are shown in Fig. 2. The experimental dependence $\tilde{f}_{rep}(T)$ can be well described by the formula (solid line on Fig. 2)

$$\tilde{f}_{rep} = \tilde{f}_{rep}(0) - 3\times 10^{-3} t^{3/2} . \tag{10}$$

The dotted line on Fig. 2 corresponds to the equation

$$\tilde{f}_{rep} = 0.15\times 10^6 (1-t^4)^{3/2} \times \frac{4-[1.23-2\ln F(T)]F^2(T)}{4F(T)} , \tag{11}$$

where $t = T/T_c$ and

$$F(T) = \frac{0.074(1-t^4)^{1/2}}{(1-t)^{1/2}} ,$$

which has been obtained by substituting Eqs. (6), (8), and (9) into Eq. (7).

Here we should note that at low temperatures the influence of geometrical factors [see Eq. (4)] on the pinning force will decrease the slope of the transition boundary to zero at $T=0$. However, in our experimental temperature window

$$5 = 0.18T/T_c \le T \le 0.82T/T_c = 23 \text{ K}$$

this effect was not observed; thus it is justified to use the linear Eq. (6) for calculations in that temperature region.

The value of the effective pinning potential barrier U_{eff} at different temperatures, which on the transition boundary equals $\tilde{U}_{rep}$, can be estimated by analogy with the estimate of $\tilde{f}_{rep}$. U_{rep}, which is responsible for repulsion of the two flux lines per unit length of the line, is given by[17]

$$U_{rep} = \frac{\phi_0 h_{12}}{4\pi} , \quad h_{12} = \frac{\phi_0 K_0(x)}{2\pi\lambda^2} , \tag{12}$$

where $K_0(x)$ is the McDonald function of zero order. For $x = d/\lambda \ll 1$ the function $K_0(x)$ may be approximated by

$$K_0(d/\lambda) = 0.11 - \ln\frac{d}{\lambda} + \frac{(1.11-\ln d/\lambda)d^2/\lambda^2}{4} . \tag{13}$$

The values of U_{eff} calculated from experimental results using Eq. (12) are shown in Fig. 3. The dotted line in Fig. 3 corresponds to the equation

$$U_{eff} = [0.11 - \ln F(T)] + \frac{1.11-\ln F(T)}{4} F^2(T) , \tag{14}$$

which has been obtained by substituting Eqs. (6), (9), and (13) into Eq. (12), and $F(T)$ is the same as in Eq. (11).

As can be seen from Fig. 3 the effective potential barrier for pinning of a single vortex on a pinning center falls almost linearly with increasing temperature, and harmonic thermal fluctuations strongly decrease the effective pinning force as $T^{3/2}$ as shown in Fig. 2.

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[1] A. I. Larkin, Zh. Eksp. Teor. Fiz. **58**, 1466 (1970) [Sov. Phys. JETP **31**, 784 (1970)].

[2] A. I. Larkin and Yu. N. Ovchinnikov, J. Low Temp. Phys. **34**, 409 (1979).

[3] C. Politis, V. Buntar, and V. P. Seminozhenko, Int. J. Mod. Phys. B **7**, 2163 (1993).

[4] V. Buntar, Phys. Lett. A **184**, 131 (1993).

[5] M. V. Feigelman and V. M. Vinokur, Phys. Rev. B **41**, 8986

(1990).

[6]D. R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988).

[7]V. V. Shmidt, *Introduction in Physics of Superconductivity (Vvedenie v Fiziku Sverhprovodnokov)* (Nauka, Moscow, 1982), p. 156.

[8]From the well-known empirical equation $H_{cm} = H_{cm}(0)[1-(T/T_c)^2]$ at low temperature $(T/T_c \ll 1)$, one can consider H_{cm} as temperature independent. The same is true for ξ.

[9]N. Ashcroft and N. Mermin, *Solid State Physics* (Mir, Moscow, 1979), Vol. II, Eq. (25.20).

[10]L. I. Glazman and A. E. Koshelev, Phys. Rev. B **43**, 2835 (1991).

[11]A. F. Hebard, P. L. Gammel, C. E. Rice, and A. F. Levi, Phys. Rev. B **40**, 5243 (1989).

[12]A. F. Hebard, A. T. Fiory, M. P. Siegal, J. M. Phillips, and R. C. Haddon, Phys. Rev. B **44**, 9753 (1991).

[13]H. Traüble and U. Essmann, J. Appl. Phys. **39**, 4052 (1968).

[14]A. A. Abrikosov, Zh. Eksp. Teor. Fiz. **32**, 1442 (1957) [Sov. Phys. JETP **5**, 1174 (1957)].

[15]C. Politis, A. I. Sokolov, and V. Buntar, Mod. Phys. Lett. B **6**, 351 (1992).

[16]Y. J. Uemura, A. Keren, L. P. Le, G. M. Luke, B. J. Sternlieb, W. D. Wu, J. B. Brewer, R. L. Whetten, S. M. Huang, S. Lin, R. B. Kaner, F. Diederich, S. Donovan, G. Grüner, and K. Holczer, Nature **352**, 605 (1991).

[17]P. G. De Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966).

[18]V. Buntar, C. Politis, and U. Eckern, Mod. Phys. Lett. B **6**, 1037 (1992).