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# A crossover in the temperature behavior of the perpendicular upper critical magnetic field of layered superconductors and thin films

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A mechanism which relates the upturn of the perpendicular upper critical magnetic field $H_{c2}^{\perp}(T)$ in layered superconductors and thin films with the structural inhomogeneity in the bulk of the sample, provided that the local critical temperature $T_c^*$ inside the inhomogeneity is higher than in the rest of the sample ($T_c$) is proposed. Within the Ginzburg-Landau approach an equation which describes two types of experimentally observed nonlinearities in $H_{c2}^{\perp}(T)$ near $T_c$ for *ISN* (insulator-superconductor-normal metal) and *NSN* layer configurations, is found. In the *NSN* case a crossover from the linear branch $H_{c2}^{\perp}(T) \propto (T_c - T)$, for fields $H \leq H_m$, to the nonlinear branch with the upturn, if $H > H_m$, takes place. The crossover field $H_m$ is inversely proportional to the local enhancement of the critical temperature $(T_c^* - T_c)$ and the distance $R$ to the surface (the nearest surface, in case of a thin film). In the *ISN* case the upturn holds for $H < H_m$, whereas for higher fields $H_{c2}^{\perp}(T)$ crosses over to the linear branch. In the *ISI* case the $H_{c2}^{\perp}(T)$ is a linear function.

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## 1. Introduction

The nonlinear behavior of the upper critical field $H_{c2}$ has been observed first in the layered dichalcogenides of transient metals in the beginning of the 1970s [1] and was given then much attention as a possible signal of non-BCS pairing in these materials. In the 1980s, an artificially prepared superconducting superlattices (SL) have been an object of intensive studies which revealed a number of nonlinearity types in the temperature behavior of the $H_{c2}(T)$: the positive curvature [an upturn of the $H_{c2}(T)$ near the critical temperature $T_c$], square-root and linear crossovers, the Takahashi-Tachiki crossover (in $S/S'$ superlattices), and the power law $H_{c2}^{\parallel} \sim [1 - (T/T_c)]^{\gamma}$ with $1/2 < \gamma < 1$ in quasi-periodic [2] and fractal [3,4] superlattices. The control over the width of layers, their number and content, as well as the deposition sequence order made it possible to clear up in detail the relationship between the structure of the artificial superlattices and the form of the $H_{c2}(T)$. A review of the temperature behavior of the upper critical field $H_{c2}(T)$ in superlattices is given in Ref. 5. The nonlinearities of the $H_{c2}(T)$ have been observed in different types of high-$T_c$ layered cuprates and superlattices made from novel materials such as YBaCuO/PrBaCuO superlattices [6].

Theoretically, the problem of calculation of $H_c(T)$ reduces to the eigenvalue problem for a fictitious particle in an external magnetic field. In the case of parallel fields this problem has been solved both numerically [7] and analytically [8–10] for different types of periodic *SI* and *SN* superlattices. A theory of the temperature dependence of the $H_{c2}^{\parallel}(T)$ for quasi-periodic SL was developed in Refs. 10,11. In contrast with the parallel field $H_{c2}^{\parallel}(T)$, where the nonlinearities are due to the lifting of degeneracy of the lowest Landau level on the orbit center position, the perpendicular field $H_{c2}^{\perp}(T)$ in all existing theories is a linear function of $T$, because the lowest Landau level in this geometry equals its standard value $\hbar\Omega/2$ ($\Omega = eH/mc$ is the cyclotron frequency). Setting this value equal to the coefficient $\alpha(T)$ of the Ginzburg-Landau expansion, we obtain the linear dependence $H_{c2}^{\perp} \sim 1 - (T/T_c)$. The exceptions are twinned crystals of the YBaCuO type and the artificial superlattices PbTe/PbS (Ref. 12) where the mismatch dislocations make a quasi-square two-dimensional lattice at the boundaries between the neighboring layers. A theory for the positive curvature of the $H_{c2}^{\parallel}$ in PbTe/PbS superlattices was given in Ref. 10. On the other hand, the positive curvature of the $H_{c2}^{\perp}(T)$ has been observed in a periodic SL [13–16], fractal SL, superconducting SL with magnetic interlayers [17], and intercalated layered crystals [18,19], including high-$T_c$ cuprates [20]. (In the latter a positive curvature close to zero temperature also has been observed [21]. We do not consider it here.) In contrast with the specific case of PbTe/PbS superlattice, in other artificially fabricated SL this nonlinearity cannot be related to some superstructure in the plane perpendicular to the external field, so that the above-mentioned mechanisms of the Landau level broadening cannot explain an upturn in the $H_{c2}^{\perp}(T)$. The positive curvature of the $H_{c2}^{\perp}(T)$ is a property inherent to all types of SL, regardless of the layer stacking sequence order. On the other hand, it seems rather sensitive to the quality of layers in SL, because an upturn in the $H_{c2}^{\perp}(T)$ was observed only in a small portion of the samples studied so far. The physical reason behind this phenomenon is not understood yet. The relationship between the quality of a layered crystal and the positive curvature of the $H_{c2}^{\perp}(T)$ has been clearly demonstrated in Refs. 18 and 19, where a positive curvature was observed after the intercalation of layered single crystals of $2H$-$NbSe_2$ by molecules of TCNQ and Sn atoms.

Very instructive observations were made in some experimental studies [4,17]. An upturn in the $H_{c2}^{\perp}(T)$ of a single Nb layer deposited on a dielectric substrate was not found in those studies [4,17], whereas it has appeared in triple layers and SLs Nb/Gd and Nb/Cu fabricated in the same series of experiments. These results show that boundary conditions at interfaces between superconducting and metal (or insulator) layers play a crucial role in physics driving the nonlinearities in the $H_{c2}^{\perp}(T)$.

In this paper we propose a mechanism of the positive curvature of the $H_{c2}^{\perp}(T)$ near $T_c$ due to the structural inhomogeneities in the bulk of a layer. This mechanism gives a qualitative description of all types of nonlinearities in the $H_{c2}^{\perp}(T)$ observed near $T_c$ in artificially fabricated SLs.

Our paper is organized as follows. In Sec. 2 the problem of calculations of the perpendicular critical field is reduced, in the adiabatic approximation, to the eigenvalue problem for a «particle» in a one-dimensional potential well which experiences an additional action from the surface. In Sec. 3 the equations for $H_{c2}^{\perp}(T)$ are derived. They describe nonlinearities in the $H_{c2}^{\perp}(T)$ of a thin film and SLs near $T_c$ with a decrease in temperature. The discussion and comparison of the results with experiments on layered superconductors are given in Sec. 4.

## 2. Formulation of the problem and the model

The problem of calculations of the upper critical field $H_{c2}^{\perp}(T)$, as is well known, reduces to the eigenvalue problem for the lowest Landau level. In the case of the Ginzburg-Landau approach an appropriate Schrödinger equation for a «particle» is

$$\hat{H}\Psi = -\,\alpha(T)\Psi \,, \qquad (1)$$

where $\Psi$ is the order parameter, and $\alpha(T)$ stands for the coefficient in front of the term $|\Psi|^2$ in the Ginzburg-Landau expansion. The physics of nonlinearities of the function $H_{c2}^{\perp}(T)$ in different types of regular and quasi-periodic superlattices is based on the fact that in these structures, due to the lift of the Landau level degeneracy on the orbit center position, the lowest edge of the energy spectrum $\epsilon_{\min}(H)$ is below $\hbar\Omega/2$. Nonlinearity of the function $\epsilon_{\min}(H)$ results then in the nonlinearity of the function $H_{c2}^{\perp}(T)$, which is a solution of the equation $\epsilon_{\min}(H_{c2}) = -\,\alpha(T)$. This approach proved to be very useful for studies of the $H_{c2}^{\parallel}(T)$, as was discussed in the previous section. In the case of a perpendicular orientation of the magnetic field, the problem of the positive curvature of the $H_{c2}^{\perp}(T)$ in thin layers and superlattices remains unsolved. The explanation of the positive curvature of the perpendicular critical fields in superlattices of the type PbTe/PbS, given in Ref. 10, is essentially based on the same idea that holds for calculations of the $H_{c2}^{\parallel}(T)$ in superlattices, because the upturn in the $H_{c2}^{\perp}(T)$ in these materials is attributable to the two-dimensional net of mismatch dislocations.

The situation with the $H_{c2}^{\perp}(T)$ is absolutely different because artificial SLs are assumed to be uniform along the layers and, hence, cannot broad the Landau levels into bands. On the other hand, among the numerous superlattices fabricated so far [5] only a few [4,13–19] have displayed a positive curvature in the $H_{c2}^{\perp}(T)$, while the great majority of them yield a linear temperature behavior near $T_c$. This linearity in $T$ is in agreement with theory, since the lowest energy level, $\epsilon_{\min}$, in this case is $\hbar\Omega/2$ and, hence, $H_{c2}^{\perp} \sim T_c - T$. Of course, perfectly uniform SL is no more than a mere theoretical model and real SLs are far from being ideal periodic structures because of uncontrollable inhomogeneities introduced during the process of their fabrication. We will show in what follows that a structural inhomogeneity of content, which locally enhances the superconductivity of a layer or thin film, gives rise to the positive curvature of $H_{c2}^{\perp}(T)$.

Let us assume that an inhomogeneity exists in a thin superconducting film at a distance $a$ from its surface, where local conditions for superconductivity are better than those in the rest of the sample, so that the local critical temperature $T_c^*$ is higher than the temperature $T_c$. In high-$T_c$ cuprates, for example, such an inhomogeneity may be due to the oxygen concentration fluctuations since the local oxygen content is a factor which mainly determines the local $T_c$. Nonuniform distribution of intercalating molecules can also cause a local enhancement of the superconductivity in intercalated layered superconductors. For simplicity we assume a cylindrical-shape inhomogeneity, so that the Schrödinger equation can be written in the symmetric gauge, $\mathbf{A} = \frac{1}{2}\,[\mathbf{Hr}]$, in the form

$$\hat{H}\Psi_E(\rho, z) = E\Psi_E(\rho, z) \ , \tag{2}$$

where

$$\hat{H} = \hat{H}_1(\rho) + \hat{H}_2(z) + U(\rho, z) \ . \tag{3}$$

Here $\hat{H}_2(z)$ is a Hamiltonian which is related to the particle motion along the field, $\boldsymbol{\rho} = (\rho, \varphi)$ are the polar coordinates in the plane perpendicular to the external field $\mathbf{H}$; $z$ is the coordinate along the field $\mathbf{H}$, and $U(\rho, z) < 0$ is a «potential well» associated with the inhomogeneity. The Hamiltonian, relevant to the motion of a «particle» in an external magnetic field in the plane, is

$$\hat{H}_1(\rho, \varphi) = -\frac{\hbar^2}{2\mu}\left[\frac{1}{\rho}\frac{\partial}{\partial\rho}\,\rho\,\frac{\partial}{\partial\rho} + \frac{1}{\rho^2}\frac{\partial^2}{\partial\varphi^2} + \frac{eH}{\hbar c}\,\hat{l}_z\right] + \frac{e^2H^2}{8mc^2}\,\rho^2 \ . \tag{4}$$

The eigenfunctions of the Hamiltonian (4) can be written in the form

$$\Psi_{Em}(\rho, \varphi) = \frac{e^{im\varphi}}{\sqrt{2\pi}}\sqrt{\rho}f(\rho) \ , \tag{5}$$

where $f(\rho)$ satisfies the equation

$$f'' + \frac{2}{\rho}f' + \left[\frac{2\mu E}{\hbar^2} + \frac{eHm}{c\hbar} - \frac{m^2 - 1/4}{\rho^2} - \frac{\rho^2}{4L^2}\right]f = 0 \ . \tag{6}$$

The solution of Eq. (6) is given by

$$\sqrt{\rho}f(\rho) = C\exp\left(\frac{-\rho^2}{4L^2}\right)\left(\frac{\rho}{L}\right)^{|m|}F\left(-n_\rho, |m| + 1, \frac{\rho^2}{2L^2}\right). \tag{7}$$

The energy spectrum is determined by the condition that the hypergeometric function $F(a, b, c)$ reduces to a polynomial, which yields

$$E_m = \hbar\Omega\left(n + \frac{1}{2}\right), \tag{8}$$

$n = n_\rho + (|m| + m)/2$ and $m = -\infty, \ldots, -1, 0, \ldots n$. Here $n$ and $n_\rho$ are integers; $\Omega = eH/\mu c$ is the cyclotron frequency, and $L = (\hbar/\mu\Omega)^{1/2}$ stands for the magnetic length.

The normalization constant $C$ in Eq. (7) is

$$C = \left[\frac{(|m| + n_\rho)!}{n_\rho!}\right]^{1/2}(|m|!L)^{-1} \ . \tag{9}$$

Since $T_c^*$ is only slightly greater than $T_c$, we can assume that influence of the potential $U(\rho, z)$ is weak compared to the action that the external field exerts on a particle. This means that an adiabatic approximation can be applied to the eigenvalue equation (2). Since we are interested in the lowest energy level $n = 0$, the adiabatic approximation in this case means that the energy and the wave function should be taken in the following approximate form [22]:

$$\Psi_{Em\epsilon}(\mathbf{r}) \approx \Psi_{0m}(\rho, \varphi)\Psi_{\epsilon m}(z) \ , \tag{10}$$

$$E = \frac{\hbar\Omega}{2} + \epsilon \tag{11}$$

where

$$\Psi_{0m}(\rho, \varphi) = \left(\frac{\rho}{\sqrt{2}L}\right)^{|m|}\exp\left(im\varphi - \frac{\rho^2}{4L^2}\right)\Big/\sqrt{2\pi|m|!}\,L \ . \tag{12}$$

Substituting (10) into Eq. (2) and eliminating $\Psi_{0m}(\rho, \varphi)$, we obtain the Schrödinger equation for the wave function $\Psi_{\epsilon m}(z)$

$$\hat{H}_2(z)\Psi_{\epsilon m}(z) + [U_{\mathrm{eff}}^m(z) - \epsilon]\Psi_{\epsilon m} = 0 \ , \tag{13}$$

where $\hat{H}_2 = -(\hbar^2/2\mu)(d^2/dz^2)$, and the effective potential energy is introduced

$$U_{\mathrm{eff}}^m(z) = \int U(\rho, z)\,|\Psi_{0m}(\boldsymbol{\rho})|^2\,d^2\boldsymbol{\rho} \ . \tag{14}$$

Since (13) is a one-dimensional Schrödinger equation, and $U_{\mathrm{eff}}^m(z)$ is negative, there should be at least one bound state in the potential well made by $U_{\mathrm{eff}}^m(z)$. In the case of boundless sample the eigenvalue $\epsilon$ in Eq. (13) is negative and strongly depends on the magnitude of the external field $H$, so that the minimal energy in the eigenvalue problem (2) in adiabatic approximation is given by

$$E_{\min} = \frac{\hbar\Omega}{2} - \epsilon_0(H) \ . \qquad (15)$$

Thus, $E_{\min}(H)$ is lower than $\hbar\Omega/2$ and, in general, is a nonlinear function of $H$, because $\epsilon_0(H)$ is a nonlinear function of $H$, as one can see from Eqs. (12) and (14), which yield the following expression for the potential well in this case:

$$U^m_{\text{eff}}(z) = \frac{2}{|m|!} \int\limits_0^\infty U(\rho, z)\tilde{\rho}^{\,2|m|+1} \exp(-\tilde{\rho}^2)\, d\tilde{\rho} \ , \qquad (16)$$

where $\tilde{\rho} = \rho/\sqrt{2}L$.

For small (compared to $\hbar\Omega$) potential $U(\rho, z)$ the one-dimensional potential well, given by Eq. (16), is shallow and $\epsilon_0(H)$ can be evaluated as [22]

$$\epsilon_0(H) \simeq \frac{mU_0^2}{2\hbar^2} \ , \qquad (17)$$

$$U_0 = \int\limits_{-\infty}^{\infty} U^0_{\text{eff}}(z)\, dz \ . \qquad (18)$$

The presence of a boundary, as is well known, can dramatically change the situation with the bound state because the value which $\Psi$ takes at the boundaries of a film strongly affects the possibility of a potential well to create a bound state. Two different types of the boundary conditions take place at the interfaces: $\Psi = 0$ for insulator-superconductor ($IS$) boundary and $d\Psi/dx = 0$ for the $NS$ boundary of the normal metal with the superconductor [23]. Thus, we have three different cases for the superconductor layer ($S$) sandwiched between the insulating ($I$) or the normal metal ($N$), which we denote as $ISI$, $ISN$, and $NSN$. The analysis given in the next section shows that conditions for the creation of a bound state of the particle in the potential well are different for these three cases.

Since the depth of a well, $U^0_{\text{eff}}(z)$, grows with the enhancement of an external field $H$, we can expect a crossover from the regime $E_{\min}(H) = \hbar\Omega/2$ to the regime where $E_{\min}(H) = \hbar\Omega/2 - \epsilon_0(H)$, when $H$ crosses over some value $H^*$. The crossover field $H^*$ corresponds to such a depth of the potential well which permits to create a bound state in the well for a given value of distance between the boundary and the well. In the context of our analysis, this crossover corresponds to the transition from the linear branch, $H^{\perp}_{c2} \propto T_c - T$, to the nonlinear branch which goes above the linear branch as the field $H$ increases to a value larger than $H^*$. The temperature dependence of the upper critical field, $H^{\perp}_{c2}(T)$, can then be determined from the equation $E_{\min}(H^{\perp}_{c2}) = -\alpha(T)$. To simplify further calculations, we will make some additional assumptions which do not change the physics beyond the above crossover. We first assume that the radius of the inhomogeneity, $R$, is less than the magnetic length, $L = (\hbar c/eH_{c2})^{1/2}$, which near $T_c$ is of the order of the coherence length $\xi(T) = \xi_0/(1 - T/T_c)^{1/2}$ because $H_{c2} = \Phi_0/2\pi\xi^2(T)$. Therefore, the condition $R << L$ reduces to the inequality $R << \xi(T)$, which is easy to satisfy near $T_c$ even for sufficiently large (in the lattice constant scale) $R$. The quantity $\Phi_0$ stands for the flux quantum. Under this condition assuming $U(\rho, z) = -|U(z)|$ if $\rho \le R$ and $U(\rho, z) = 0$ otherwise, we have from Eq. (16)

$$U^0_{\text{eff}} = -|U(z)|\, I(H) \ , \qquad (19)$$

where

$$I(H) \approx \frac{2\pi R^2 H}{\Phi_0} \ . \qquad (20)$$

Thus, the effective potential well depth is proportional to the flux, $\Phi = 2\pi R^2 H$,

$$U^0_{\text{eff}} = -|U(z)|\frac{\Phi}{\Phi_0} \ . \qquad (21)$$

Since the precise form of the potential well is unknown, we will simulate it, as is generally accepted, with the $\delta$-well:

$$U^0_{\text{eff}} = -V\frac{\Phi}{\Phi_0}\,\delta(z) \ , \qquad (22)$$

where

$$V = \int\limits_{-\infty}^{\infty} dz\, |U(z)| \ .$$

In the particular case of a layered superconductor such as $NbSe_2$ or the one from the family of high-$T_c$ cuprates, the approximation given by Eq. (22) is quite acceptable because the inhomogeneity that belongs to a certain layer has a form of a «pancake». Such «pancakes» may be due to the nonuniform distribution of intercalating molecules in dichalcogenides of transient metals or oxygen (in the case of cuprates) since local concentration of these elements determines the local value of the critical temperature $T_c$ .

## 3. Analytic consideration of the positive curvature and crossover of the upper critical field $H^{\perp}_{c2}(T)$

Consider a thin superconducting film of thickness $d$, which amounts to a few $\xi$ or less and which contains an inhomogeneity with the effective potential (22) located at a distance $a$ from the surface. The problem of calculation of the $H^{\perp}_{c2}(T)$ then reduces to finding the lowest eigenvalue of the Schrödinger equation (13) with

$$U^0_{\rm eff} = -V\frac{\Phi}{\Phi_0}\,\delta(z-a) \tag{23}$$

and appropriate boundary conditions. We first consider the case of $NSN$ sandwich, for which the boundary conditions are $\Psi(0) = \Psi(d) = 0$. To satisfy these condition, we write the solution in the form

$$\Psi_1(x) = A\sinh \kappa x\ , \quad \text{for } 0 \le x \le a\ ,$$
$$\Psi_2(x) = B\sinh \kappa(x-d)\ , \quad \text{for } a \le x \le d\ . \tag{24}$$

Here $\kappa^2 = 2m|\epsilon|/\hbar^2$ . The constants $A$ and $B$ can be found from the corresponding boundary conditions at the $\delta$-well

$$\Psi_2'(a) - \Psi_1'(a) = -\frac{2mV\Phi}{\hbar\Phi_0}\Psi_1(a)\ ,$$
$$\Psi_1(a) = \Psi_2(a) \tag{25}$$

It follows immediately from Eqs. (24) and (25) that the energy of the bound state $\epsilon_0$ is determined by the only root of the equation

$$\frac{H}{H_m} = YF(Y)\ , \tag{26}$$

where

$$F(Y) = \coth Y + \coth(Yb/a)\ . \tag{27}$$

The energy of the bound state is then given by

$$\epsilon_0 = -\frac{\hbar^2Y^2}{2ma^2}\ . \tag{28}$$

We have assumed here for certainty that $b = d - a > a$. In the opposite case $a$ should be replaced by $b$. It is easy to see that the eigenvalue equation (26) has a solution only if $H > H^*$, where

$$H^* = H_m\left(1 + \frac{a}{b}\right), \tag{29}$$

and $H_m$ is a threshold field given by

$$H_m = \frac{\Phi_0\hbar^2}{\pi R^2 maV}\ . \tag{30}$$

A sample which occupies a half-space corresponds to the limit $b \to \infty$ in Eqs. (27)–(29). We thus obtain the following picture. If $H \le H_m$ , the lowest eigenvalue of the problem under study is $E_{\min} = \hbar\Omega/2$. For fields $H > H_m$ the minimal energy is $E_{\min} = \hbar\Omega/2 - \epsilon_0(H)$. Equating then $E_{\min}$ to the Ginzburg-Landau coefficient $\alpha$, we have

$$H^{\perp}_{c2} = H(0)\left(1 - \frac{T}{T_c}\right), \quad \text{if } H \le H^*\ , \tag{31}$$

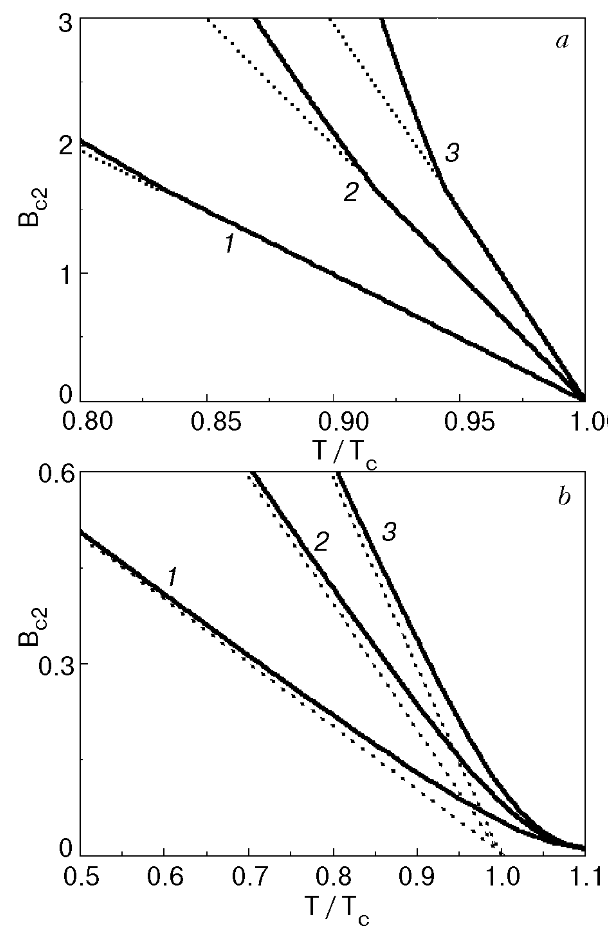


*Fig. 1*. The dependence of the reduced perpendicular critical magnetic field $B_{c2}$ on the temperature $T/T_c$ for the $NSN$ ($a$) and $ISN$ ($b$) cases calculated from the Eq. (26) with: ($a$) $F(Y)$ given by Eq. (27), $W = (\xi/a)^2 = 0.1$ and $G = 10;\ 20;\ 30$ (curves *1, 2, 3* ); ($b$) $F(Y)$ given by Eq. (34) $W = 0.1$ and $G = 1;\ 2;\ 3,$ (curves *1, 2, 3*). The parameter $G = H(0)/H_m$ and the ratio $b/a = 1.5$ for both cases.

$$H_{c2}^{\perp} = H(0)\left(1 - \frac{T}{T_c}\right) + Y^2 H(0)\left(\frac{\xi}{a}\right)^2 , \quad \text{if } H > H^* , \tag{32}$$

where $H(0) = \Phi_0/(2\pi a^2)$, and $Y(H/H_m)$ is determined by Eq. (26). We see that for $H \le H_m$ the perpendicular critical field, given by Eq. (31), is a linear function of temperature $T$ and at $H = H_m$ it crosses over to the nonlinear branch of Eq. (32), which goes higher than (31) and has an upturn or a so-called «positive curvature». The dependence of the reduced critical field, $B_{c2} = H_{c2}^{\perp}/H_m$ , on the reduced temperature $T/T_c$ for different values of the parameters $G = H(0)/H_m$ and $W = (\xi/a)^2$ is shown in Fig. 1.

Since Eq. (13) is valid for

$$|\epsilon_0| << \frac{\hbar\Omega}{2} , \tag{33}$$

the second term on the right-hand side of Eq. (32) should be small compared to the first term. This condition determines the formal validity of Eq. (32). It follows then from Eq. (32) that the smallness of the parameter $W \propto a^{-2}$ is favorable for the applicability condition (33). On the other hand, $H_m \propto a^{-1}$, so values of W and $H_m$ decrease with increasing of the separation between the surface and the inhomogeneity.

In the case of a film sandwiched between an insulating and a normal-metal (or ferromagnet) layer, i.e., in the *ISN* case, the boundary conditions are $d\Psi(0)/dx = 0$ and $\Psi(L) = 0$. The appropriate function $F(Y)$ in Eq. (26) is

$$F(Y) = \coth Y - \tanh\left(Y \frac{b}{a}\right). \tag{34}$$

The solution of Eq. (26), with the $F(Y)$ given by Eq. (34), yields a nonzero root only for $H < H_m$ . This means that a crossover in $H_{c2}^{\perp}(T)$ for the *ISN* case is somewhat different from that we have described above for the *NSN* sandwich: an upward-like branch (32) for $H < H^*$ crosses over to the linear branch (31) when the field exceeds the value $H_*$ . The results of a numerical analysis for the $H_{c2}^{\perp}(T)$ in the *ISN* case is shown in Fig. 1,*b*.

In the *ISI* case the function $F(Y)$ is determined by the Schrödinger equation (13) and by the boundary conditions $d\Psi(0)/dx = d\Psi(L)/dx = 0$, which yield

$$F(Y) = \tanh Y - \tanh\left(Y \frac{b}{a}\right). \tag{35}$$

Substitution of Eq. (35) into Eq. (26) gives an equation which has no solution for positive $Y$. This means that $\epsilon_0 = 0$ and $H_{c2}^{\perp}(T)$ for the *ISI* sandwich is given by the linear function (31) in accordance with the experiments [4,17]. In the next section we will discuss the physical meaning of the results obtained in the context of a current experimental situation in the field.

## 4. Discussion and conclusions

Let us summarize the results obtained in the previous sections from the viewpoint of their relevance to experiments done so far. We see that at least two major preconditions are necessary for deviation of the $H_{c2}^{\perp}(T)$ from the linear behavior (31): a) structural inhomogeneities with local enhancement of the critical temperature and b) an appropriate boundary condition of the *NSN* or *ISN* type. Therefore, structurally perfect films and multilayers should not display nonlinearities of the $H_{c2}^{\perp}(T)$ near $T_c$ . This assertion is in agreement with the fact that an upturn in $H_{c2}^{\perp}(T)$ has been observed only in limited number of experiments on different superconducting SLs, whereas the rest of them show linear behavior of the perpendicular critical field [5]. But even the prerequisite a) is satisfied in a single film; it does not display a nonlinearity in $H_{c2}^{\perp}(T)$ when sandwiched between the insulators, i.e., in the *ISI* case. This conclusion of our theory is confirmed by experiments reported elsewhere [4,17]. Those experiments showed that the $H_{c2}^{\perp}(T)$ of a single Nb film deposited on a dielectric substrate in vacuum is a linear function of $T$ near the phase transition, but it becomes upturned in triple layers and multilayers Nb/Gd (Ref. 17) and Nb/Cu (Ref. 4) fabricated from the Nb films. The latter, as was found in Nb/Gd and Nb/Cu superlattices, have a grained structure necessary for our approach. Thus, we can explain the above experimental observations as follows. A single layer deposited on an insulating sapphire substrate in vacuum belongs to the *ISI* case in our classification and, hence, has no nonlinearities in the $H_{c2}^{\perp}(T)$ behavior. The situation changes in the case of triple layers Nb/Cu/Nb and Nb/Gd/Nb, because they are of the *ISN* type (since each of the two Nb layers in the triple layer makes contact with one insulator and one normal metal layer) and should display a crossover of the kind shown in Fig. 1,*b*. The multilayers Nb/Cu and Nb/Gd are of the NSN type in the bulk of the sample and of the *ISN* type for the marginal layers at the top and the bottom of a SL (where the superconductor layer contacts either with the vacuum or with an insulating substrate). Therefore, the nonlinearity (see Fig. 1) which displays a specific SL depends on which of its layers (marginal or the one in the bulk of a SL) yields the largest $H_{c2}^{\perp}(T)$.

The intercalation of a layered crystal $NbSe_2$, as was shown in Refs. 18 and 19 also gives rise to the upturn in the $H_{c2}^{\perp}(T)$. Let us consider in a more detail the case reported in Ref. 18, where the temperature behavior of the $H_{c2}^{\perp}(T)$ of layered single crystals 2$H$-$NbSe_2$, intercalated by molecules of TCNQ, has been studied. Before the intercalation, the $H_{c2}^{\perp}(T)$ was found to be a linear function of the temperature. After the intercalation, the $H_{c2}^{\perp}(T)$ became a nonlinear function, whose shape near $T_c$ reported in Ref. 18 is as follows: a linear branch up to $H_m \approx 0.8$ T and then a smooth upturn with further decrease in temperature. The critical temperature of intercalated 2$H$-$NbSe_2$, $T_c^* = 6.5$ K, is lower than that of a nonintercalated crystal, where $T_c = 7.2$ K. The physical reason behind the lowering of $T_c$ after the intercalation is that molecules of the TCNQ, when placed between the superconducting sheets, diminish the concentration of electrons in them since the TCNQ is a very active acceptor. On the other hand, the intercalation procedure cannot provide a perfectly uniform distribution of the TCNQ molecules across the sample. The latter means that after the intercalation some inhomogeneities must inevitably appear with the lower local concentrations of the TCNQ molecules. The corresponding local critical temperatures in them are higher than in the rest of the sample. The above data allow us to estimate the local enhancement as $\Delta T_c = T_c - T_c^* \approx 0.7$ K $<< T_c$. Therefore, the value of $V$ in Eq. (30) can be taken as $V = \Delta T_c\, d$, where $d$ is of the order of the distance between the superconducting sheets in the intercalated 2$H$-$NbSe_2$. Assuming $d \approx 10$ Å, $a \approx 10$–$100$ Å, and $H_m \approx 1$ T, we can estimate $R$ from Eq. (28) as $R \approx 10^3$–$10^4$ Å, which seems plausible for the reported [18] concentrations of the TCNQ molecules in the intercalated 2$H$-$NbSe_2$. One can check the above theoretical model by measurements of the $H_{c2}^{\perp}(T)$ and by concurrent control of the spatial distribution of the TCNQ molecules at different stages of the intercalation.

It is rather tempting to apply our model to another yet unresolved problem — nonlinearity of the $H_{c2}^{\perp}(T)$ near $T_c$ in layered high-$T_c$ cuprates. In these materials the oxygen is the agent which controls the local values of the critical temperature. Thus, spatial fluctuations of the oxygen in the plane would result in «pancakes» where the local $T_c$ is higher than in the rest of a sample. According to the previous consideration, such a type of inhomogeneity is a prerequisite for the positive curvature of the $H_{c2}^{\perp}(T)$. Although an upturn in the $H_{c2}^{\perp}(T)$ near $T_c$ in layered high-$T_c$ cuprates has been reported in many publications, it is well known that to measure this quantity in detail is very difficult in these materials because of the resistive transition broadening in an external magnetic field [20]. In contrast, the melting line $B_m(t)$ is a much better measurable quantity in high-$T_c$ cuprates. Its shape via the elastic moduli, $c_{i,j} = c_{i,j}(b)$, depends on the $H_{c2}(T)$, where $b = H/H_{c2}(T)$. Therefore, a crossover in the line $H_{c2}(T)$ inevitably should manifest itself in the form of the function $B_m(T)$. This rather evident fact, as was shown in Ref. 24, must be taken into account in calculating the shape of the melting line $B_m(T)$.

Consider now briefly a defect which extends through the bulk of a layered crystal. In this case the potential $U_{\text{eff}}^0(z)$ in the eigenvalue problem of Eq. (13) is given by the infinite set of periodic potential wells of the form

$$U_{\text{eff}}^0(z) = -\frac{\Phi}{\Phi_0} V \sum_n \delta(z - an) , \qquad (36)$$

where $a$ is the interlayer spacing. The eigenvalue problem of Eq. (13) is now exactly the well-known Kronig—Penny model, whose lowest energy level $E_{\min} = -(\hbar^2 Y^2)/(2ma^2)$ is given by the solution of the equation

$$\cosh Y - \frac{H}{H_m Y} \sinh Y = 1 . \qquad (37)$$

The critical field is determined by Eq. (32) which describes the curve that gradually upturns with a decrease in temperature from the point of $T_c$. The analytic solutions can be easily found for two cases. Near the critical temperature, i.e., for $H << H_m$ the $H_{c2}^{\perp}(T)$ is a linear function of the temperature:

$$H_{c2}^{\perp}(T) \approx H(0) \left[ 1 + 2\, \frac{H(0)}{H_m} \left( \frac{\xi}{a} \right)^2 \right] \left( 1 - \frac{T}{T_c} \right).$$

When $H >> H_m$, which corresponds to lower temperatures, and if the additional condition $\xi << a$ is satisfied, which implies that $[H\xi/(H_m a)]^2 << 1$ (the latter is the case, for example, in some highly anisotropic high-$T_c$ cuprates), the upper critical field is given by

$$H_{c2}^{\perp}(T) \approx \left( \frac{H(T)\xi}{H_m\, a} \right)^2 H(0) \left( 1 - 2e^{-H(T)/H_m} \right) + H(T) .$$

Here $H(T)$ equals to the right-hand side of Eq. (31), which is a linear function of the temperature and, hence, the $H_{c2}^{\perp}(T)$ experiences an upturn known also in the literature as the positive curvature.

In summary, we conclude that the presence of a particular type of inhomogeneity in thin films and layered superconductors, which enhances the local value of the critical temperature, is one of the physical reasons beyond the positive curvature in the temperature behavior of the $H_{c2}^{\perp}(T)$ observed in some multilayers near $T_c$ .

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