a growth rate for benzene of about 3×10^7 sec⁻¹. In summary, it has been demonstrated that

the light-medium coupling can lead to rapidly growing instabilities, and hence to large density fluctuations. Any small-scale structure in the laser beam, either present initially or caused by inhomogeneities in the medium, will thereby be enhanced, resulting in large intensity changes and so in anomalous gain.

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EFFECT OF SPIN-ORBIT SCATTERING ON THE UPPER CRITICAL FIELD OF HIGH-FIELD SUPERCONDUCTORS

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We present experimental evidence for the effect of spin-orbit scattering on the bulk upper critical field, H_{c2} , of high-field superconductors. We have measured the temperature variation of H_{c2} for three concentrated titanium alloys, viz. Ti-58 at.% V, Ti-44 at.% Nb, and Ti-52 at.% Ta. For each of these alloys the Pauli spin paramagnetism (PSP) should limit H_{c2} to a value lower than that predicted by the Ginzburg-Landau-Abrikosov-Gor'kov (GLAG) theory. It is found, however, that the measured values of H_{c2} are substantially higher than those predicted by Maki's theory¹ for the effects of the PSP on H_{c2} . More significantly, the observed deviation of the data from Maki's predictions shows a systematic trend, namely, the higher the atomic number (Z) of the column-V constituent $[V(23) \rightarrow Nb(41) \rightarrow Ta(73)]$, the greater is the deviation. This trend is the first evidence to support the recent theoretical conjecture^{2,3} that spin-orbit scattering counteracts the effect of PSP on H_{C2} . Values of the spin-orbit scattering times, deduced from the data using the theory of Werthamer, Helfand, and Hohenberg² (WHH), are found to decrease rapidly with increasing Z, as expected,⁴ and are also in order-of-magnitude agreement with theoretical estimates.

It has been pointed out independently by Clogston⁵ and by Chandrasekhar⁶ that the PSP places an upper limit on H_{C2} . Subsequently Maki¹ showed, by a detailed calculation, that the PSP lowers the upper critical field at temperature T from the GLAG value $H_{c2}^{*}(T)$ to a value $H_{c2}(T)$, and that it also modifies the temperature variation of H_{c2} . Kim, Hempstead, and Strnad⁷ and the present authors^{8,9} have shown experimentally that while $H_{c2}(0)$ is lower than $H_{c2}^{*}(0)$, it is higher than predicted by Maki's theory. Moreover, it was found^{8,9} that the effect of the **PSP** on the temperature variation of H_{c2} is smaller than predicted by Maki. To explain these findings WHH² and Maki³ have suggested that spin-orbit scattering, which was not included in Maki's original treatment,¹ must be considered. In particular, they have shown theoretically that spin-orbit scattering counteracts the effects of PSP on H_{c2} . The present work was undertaken for the purpose of testing the validity of this explanation. In the theories of WHH and Maki, the spin-orbit scattering is characterized by a parameter which involves the spin-orbit scattering time τ_s . Since the magnitude of τ_s is not known accurately, it is difficult to compare experimental results on any <u>one</u> particular alloy with theory. On the other hand, because the spinorbit scattering is expected to increase rapidly with atomic number,⁴ we have chosen to test the theory by studying <u>several</u> alloys with widely different Z.

In Maki's theory the relative importance of the PSP is characterized by the parameter α defined as

$$\alpha = \sqrt{2H} c 2^{*}(0)/H_{p}, \qquad (1)$$

where

$$H_p = 18\,400T_C \,\mathrm{G},$$
 (2)

and T_c is the transition temperature in °K. In the limit of short electronic mean free path (dirty limit), $H_{c,2}$ *(0) is given by²

$$H_{c2}^{*}(0) = -0.69(dH_{c2}/dt)_{t=1} \equiv 0.69H_{0}, \qquad (3)$$

where $t = T/T_c$. In the present work the parameter α has been determined experimentally from the measured values of T_c and H_0 by using Eqs. (1)-(3). Alternatively, when the electronic specific-heat coefficient γ of the normal state is known,¹⁰ the field $H_{c2}^*(0)$ can be determined from the expression⁷

$$H_{c2}^{*}(0) = 3.1 \times 10^4 \rho_n \gamma T_c G,$$
 (4)

where ρ_n is the normal-state dc resistivity



FIG. 1. Temperature variation of H_{C2} . The values of α are calculated from Eqs. (1)-(3) and the measured values of T_C and H_0 .

in Ω cm and γ is in erg deg⁻² cm⁻³. This gives

$$\alpha = 2.4 \rho_{\gamma} \gamma. \tag{5}$$

According to Maki, in the absence of spin-orbit scattering, the reduced field $h(t) = H_{c2}(t)/H_0$ decreases with increasing α . In particular,

$$h(0) = 0.69(1 + \alpha^2)^{-1/2}.$$
 (6)

To describe the effects of spin-orbit scattering, WHH have introduced the parameter $\lambda_{\rm SO}$ defined as

$$\lambda_{\rm SO} = \hbar/3\pi k T_{\rm c} \tau \,. \tag{7}$$

In their theory the reduced field h(t) is (in the dirty limit) a unique function of α , λ_{SO} , and t. Qualitatively, h(t) increases with increasing λ_{SO} .

Experiments were performed on vacuumannealed specimens of Ti-58 at.% V, Ti-44 at.% Nb, and Ti-52 at.% Ta. The variation of the electrical resistance of these alloys with magnetic field intensity H was measured over the temperature interval $1.3^{\circ}K \leq T \leq T_c$. The current through the sample was always transverse to the applied magnetic field which was produced by a solenoid capable of generating a steady field up to 150 kG. With low current densities $(J \sim 0.1-30 \text{ A/cm}^2)$ the superconducting-to-normal resistive transitions were sharp (2-5 kG wide). For a given current density J, the field $H_{\gamma}(J)$ at which the resistance was equal to half its normal value was defined as the resistive transition field. In general, $H_{\gamma}(J)$ was found to decrease slightly with increasing J, and the field $H_{\gamma}(0)$ was obtained by extrapolating $H_{\gamma}(J)$ to zero current. We chose to identify H_{c2} with $H_{\gamma}(0)$.¹¹

The temperature variation of H_{c2} for the three alloys is shown in Fig. 1. For comparison with theory it is preferable to plot h(t) vs t. Such plots are shown in Fig. 2 together with the temperature dependence expected from Maki's original theory ($\lambda_{SO} = 0$) for $\alpha = 0$, 0.8, and 1.5. It is apparent from Fig. 2 that h(t) is different for the three alloys.

Examination of Table I, which lists the physical properties of the specimens, indicates that the three alloys have comparable values of $H_{C2}*(0)$, α , ρ_n , and T_c . In particular, since α does not vary appreciably from alloy to alloy, the marked variation of h(t) from one material to another cannot be attributed solely to the effect of PSP. Comparison of the data



FIG. 2. Variation of the reduced field $h = H_{C2}/H_0$ with reduced temperature *t*. Data points for t > 0.9 have been omitted. The dashed curves are taken from Maki's theory with $\lambda_{SO} = 0$. The solid curves are from the WHH theory.

in Fig. 2 with Maki's curve for $\alpha = 0$ (no PSP) indicates that the PSP affects the magnitude and temperature dependence of h(t) for Ti-58 at.% V and Ti-44 at.% Nb. However, the deviation of the data points from the curve $\alpha = 0$ is smaller, in all cases, than predicted by Maki's original theory¹ which neglects spinorbit scattering. It is also clear that this deviation is largest for Ti-58 at.% V and smallest for Ti-52 at.% Ta, which is precisely what is anticipated if the spin-orbit scattering, whose effect increases rapidly with increasing Z, counteracts the influence of the PSP on H_{C2} .

A more quantitative analysis was made by fitting the data in Fig. 2 to the WHH theory.

Table I. Physical properties of concentrated Ti alloys.

	Т _с (°К)	$(\mu\Omega \text{ cm})^{\rho} a$	<i>H</i> ₀ (kG)	$\frac{H_{c2}^{*}(0)}{(kG)b}$	αc
Ti-58 V	7.52	59	221 ± 8	153 ± 6	1.56
Ti-44 Nb	8.99	53	227 ± 16	157 ± 11	1.34
Ti-52 Ta	7.86	51	194 ± 5	134 ± 3	1.31

 $^{a}_{4.2}$ K, $H > H_{c2}$

^bFrom Eq. (3).

^cFrom Eqs. (1)-(3).

We have chosen to regard α as a fixed parameter determined by Eqs. (1)-(3) and the measured values of T_c and H_0 . On the other hand, λ_{SO} was adjusted to give the best agreement between theory and experiment.¹² For Ti-58 at.% V the best fit to the data is obtained with $\lambda_{SO} = 0.7$, while for the Ti-44 at.% Nb data, $\lambda_{SO} \approx 4.5$. These theoretical fits are shown in Fig. 2 by the solid curves. In the case of Ti-52 at.% Ta, the data points lie slightly above Maki's curve with $\alpha = 0$ (which also is the curve for any finite α when $\lambda_{SO} = \infty$). As a consequence it is impossible to obtain a good fit to the data with any value of λ_{SO} ,¹³ although the best fit is with $\lambda_{SO} = \infty$. Examination of the values of λ_{SO} for the three alloys shows that it increases rapidly with atomic number, as expected. The spin-orbit scattering time τ_s can be estimated from the best value of λ_{so} using Eq. (7). In this fashion one obtains $\tau_{s} \approx 1.5 \times 10^{-13}$ sec for Ti-58 at.% V, and $\tau_{\rm S} \approx 2 \times 10^{-14}$ sec for Ti-44 at.% Nb. From the Abrikosov and Gor'kov⁴ treatment of spin-orbit scattering, one expects that the order of magnitude of $\tau_{\rm S}$ is given by

$$\tau/\tau_{s} \sim (Ze^{2}/\hbar c)^{4}, \qquad (8)$$

where τ^{-1} is the total collision frequency of the electron and we have assumed $\tau_S \gg \tau$. For the specimens used in the present work we estimate $\tau \sim 10^{-15}$ sec. Equation (8) then gives $\tau_S \sim 10^{-12}$ sec for Ti-58 at.% V and $\tau_S \sim 10^{-13}$ sec for Ti-44 at.% Nb. These estimates agree to within an order of magnitude with the values of τ_S deduced from the observed temperature variation of H_{C2} using the WHH theory.

From a technological point of view, the present work suggests that the limitations^{5,6} imposed by the **PSP** on the generation of high magnetic fields by superconductors may be overcome by using suitable high-Z alloys.

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¹¹This choice is somewhat arbitrary inasmuch as the

resistive transition has a finite width. However, because the resistive transition is rather narrow and because the width of the transition does not depend strongly on temperature, any other reasonable choice of H_{C2} (say, the onset of resistivity) would not change the main features and conclusions of the present work.

¹²The parameter λ_{SO} can be determined most accurately when α is large and λ_{SO} is small, as is the case for Ti-58% V. For Ti-44% Nb the data indicate that $\lambda_{SO} > 2$, with a best value of $\lambda_{SO} \approx 4.5$. This result differs from the value $\lambda_{SO} = 1.5$ obtained by WHH,² who have fitted our data to their theory. This apparent discrepancy arose because in converting the measured H_{C2} to h, WHH used a value for H_0 which is higher than the average one. In addition, these authors calculated α from Eq. (5) using an estimated value for γ . The conclusion that λ_{SO} for Ti-44% Nb is significantly larger than for Ti-58% V is valid, however, in any case.

¹³There are several possible reasons for this discrepancy: (a) Effects of finite mean free path tend to increase h; (b) strong coupling effects tend to increase h; (c) if one identifies H_{c2} with the field at which the onset of resistance (at low current densities) takes place, rather than with $H_{r}(0)$, one obtains slightly lower values of h.

LIGHT SCATTERING BY SPIN WAVES IN FeF2

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The optical properties of spin waves (magnons) in the transition metal fluorides have recently received considerable attention. Most recently the direct absorption of infrared radiation by two magnons,^{1,2} as well as magnon sidebands of optical absorptions, have been observed.³⁻⁶ In addition it has been proposed that magnons might exhibit a Raman effect.^{7,8} We report here the first observation of light scattering by magnons in both first and second order. We identify the first-order, or onemagnon, scattering by the magnitude and temperature dependence of the frequency shift of the scattered light, by the polarization selection rules observed to govern the scattering, and most strikingly, by the disappearance of the scattered light when the sample temperature is raised above the Néel temperature. The identification of the second-order, or twomagnon, scattering is similar, though not quite so definite, because of possible interactions with low-frequency optical phonons. We shall discuss the two scattering processes in turn.

In these experiments the sample, a $5 \times 5 \times 7$ mm³ oriented single crystal of FeF₂,⁹ is illuminated with ~50 mW of linearly polarized, 4880-Å light from an argon ion laser. Cooling is achieved by flowing He gas over the sample at a rate determined by a feedback system containing the carbon resistor which monitors the sample temperature. In this way the sample temperature is maintained to within 0.5°K of a desired value above 10°K. Light scattered through 90° is passed through a Czerny-Turner double monochromator onto a cooled S-11 photomultiplier. The photomultiplier output is then amplified and displayed on a chart recorder ¹⁰

FeF₂ has the rutile structure¹¹ (D_{4h}) and becomes antiferromagnetic for $T < T_N = 78.5^{\circ}$ K. Although the magnon dispersion curve has not been measured, it is known from antiferromagnetic resonance (AFMR) experiments¹² that the frequency of a zone-center (k = 0) magnon at T = 0 is 52.7 cm⁻¹. One may then estimate¹ the zone-edge magnon frequency to be ~77 cm⁻¹.