## de Haas-van Alphen effect in the vortex state of Nb<sub>3</sub>Sn

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de Haas-van Alphen (dHvA) oscillations corresponding to five extremal areas of the Fermi surface have been observed in the vortex state of Nb<sub>3</sub>Sn, in magnetic fields as low as  $\sim 0.5 B_{c2}$ . Both the dHvA frequencies and effective masses are unchanged from their values in the normal state, although the quantum oscillations experience a field-dependent additional damping in the superconducting state. The results show that the Landau quantization of quasiparticles persists in the mixed state of a clean type-II superconductor.

It is well known that the Landau quantization of the motion of electrons in a magnetic field leads to an oscillatory contribution to the free energy of a metal<sup>1</sup> and, hence, an oscillatory contribution to the magnetization: the de Haas-van Alphen (dHvA) effect. Graebner and Robbins<sup>2</sup> showed that quantum oscillations (in this case magnetothermal oscillations) can persist into the mixed state of a type-II superconductor thus demonstrating the existence of Landau quantization in the superconducting state. While there is theoretical debate<sup>7</sup> about the detailed nature of the electronic excitations in the mixed state, it is clear that the dHvA effect offers a potentially unique probe of the anisotropies and nature of the quasi-particles in a superconductor.<sup>7-13</sup> Observations of small portions of the Fermi surface have recently been reported in a number of superconductors. 13-17 In this paper we describe a study of the de Haas-van Alphen effect in the normal and vortex states of the A15 compound Nb<sub>3</sub>Sn. We resolved seven dHvA frequencies corresponding to five extremal Fermi-surface orbits (and two harmonics). On entering the superconducting state, both the dHvA frequencies and effective masses are unchanged from their values in the normal state. However, the amplitude of all frequency components is diminished. We interpret our measurements within a picture where quasiparticles exist in the mixed state and compare results with recent measurements on V<sub>3</sub>Si.<sup>17</sup>

The A15 type-II superconductor, Nb<sub>3</sub>Sn, is of both theoretical and technological interest. Often cited as an example of a strong-coupling phonon-mediated BCS superconductor, its electronic structure has been the subject of extensive calculation<sup>18</sup> yielding results at the Fermi energy in satisfactory agreement with previous dHvA experiments<sup>18</sup> in the normal state. The sample of Nb<sub>3</sub>Sn used for these experiments was the same as that (sample B) used previously by Wolfrat et al. 18 for their measurements of the dHvA effect in the normal state. The crystal was grown by a chemical transport method, effected by iodine in a closed ampoule, using as starting materials 99.9% Nb and 99.9995% Sn. Its residual resistance ratio was measured to be  $\rho(T=300 \text{ K})/\rho(T\rightarrow 0)=70\pm 5$ ,  $\rho(T\rightarrow 0)=15 \text{ n}\Omega \text{ m}$ , with a superconducting critical temperature  $T_c = 18.3$  K and  $B_{c2} = 19.7$  T. Nb<sub>3</sub>Sn undergoes a martensitic transformation near 50 K from a cubic to a tetragonal structure and Wolfrat et al. concluded that their dHvA spectra in the normal state could be matched reasonably well to the calculated electronic structure of tetragonal Nb<sub>3</sub>Sn, specifically to the lighter mass sheets of the Fermi surface centered at the M and R points of the tetragonal Brillouin zone.

The present experiments were performed in pulsed magnetic fields extending to 40 T and at temperatures in the range 1.3-2.2 K. High sensitivities were achieved by using high rates of change of magnetic field ( $\sim 10^4$  T s<sup>-1</sup>), while eddy-current heating in the normal state was avoided by slotting the sample to restrict current loops to dimensions  $\leq 100 \,\mu\text{m}$ . An example of an experimental record is shown in Fig. 1(a) in which quantum oscillations are seen at fields both above and substantially below  $B_{c2}$ . Just below  $B_{c2}$  is a disturbed region of the record (shown dotted) which is thought to arise from flux redistribution. Spectral analysis of the record in Fig. 1(a), for the normal and superconducting regimes separately, is depicted in Fig. 2 by the broken and solid lines, respectively. It reveals the presence of at least seven dHvA frequencies. de Haas-van Alphen data are conventionally interpreted using the semiclassical Lifshitz-Kosevich (LK) theory for a collection of weakly interacting quasiparticles. Within this theory, each extremal crosssectional area A of the Fermi surface gives an oscillatory contribution to the magnetization,

$$\widetilde{M} \propto \alpha(B_0, T) \sin \left[ \frac{2\pi F}{B_0} + \phi \right] , \qquad (1)$$

$$\alpha(B_0, T) = B_0^{-1/2} \frac{T}{\sinh(2\pi^2 k_B T / \hbar \omega_c)}$$

$$\times \cos \left[ \frac{\pi g m^*}{2m_e} \right] R_0 R_S , \qquad (2)$$

where  $F = (\hbar/2\pi e) A$ ,  $\omega_c = eB_0/m^*$ .  $R_0 = \exp(-\pi/2\pi e)$  $\omega_c \tau_0$ ) accounts for the scattering due to static defects and

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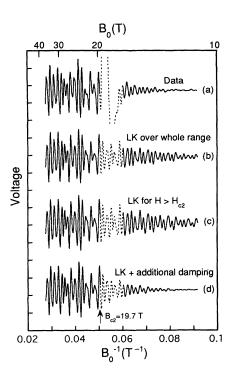


FIG. 1. (a) dHvA oscillations in Nb<sub>3</sub>Sn at T=1.3 K. The field was directed along the [001] axis of the room-temperature cubic phase. The traces are the oscillatory voltages in the pickup coil and are therefore proportional to  $\partial M/\partial t$ . Quantum oscillations are seen both above and below the critical field  $B_{c2}$  = 19.7 T. The disturbed region of the record, shown dotted, is presumed to be associated with the creation and destruction of the flux lattice and is not used in the subsequent analysis. (b) A least-squares fit to the experimental trace (a) to the Lifshitz and Kosevich (LK) theory for a collection of weakly interacting Fermions over the entire field range. The measured signal is  $\partial M/\partial t = (\partial M/\partial B)(\partial B/\partial t)$ ,  $\partial M/\partial B$  is obtained from the LK formula and  $\partial B/\partial t$  is determined from a field measuring coil. (c) A least-squares fit of conventional LK theory to the data in the normal state only, which is then extrapolated into the superconducting state. (d) A least-squares fit of LK theory to the data in the normal state is modified by the inclusion of an additional field-dependent damping term, in the vortex state (see text).

impurities,  $\tau_0^{-1}$  being the quasiparticle scattering rate, and  $R_S=1$  in the normal state. The field and temperature dependencies of  $\alpha(B_0,T)$ , for  $B_0>B_{c2}$ , are used to obtain the effective masses and scattering rates associated with the various orbits in the normal state. The results are summarized in Table I and are in good agreement with previous investigations of the normal state. <sup>18</sup>

In analyzing the data in the mixed state, it is instructive to see to what extent it can be described by the conventional semiclassical Lifshitz-Kosevich theory. Fig. 1(b) shows a least-squares fit of the LK theory  $(R_S = 1)$  to the experimental trace in Fig. 1(a) which has been made

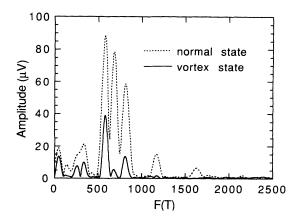


FIG. 2. Fourier (amplitude) spectrum of the dHvA oscillations, in Nb<sub>3</sub>Sn [see Fig. 1(a)]. The Fourier spectrum (with respect to  $1/B_0$ ) of the signal in the pickup coil. Broken line is for the normal state  $19.7 < B_0 < 36.4$  T and the solid line for the mixed state  $12.3 < B_0 < 17.2$  T. The spectra reveals the presence of at least seven frequencies given in Table I.

over the entire field range using the effective masses determined from the temperature dependence of the normalstate data, all other parameters have been allowed to vary. Comparing Figs. 1(a) and 1(b), it is clear that the fit is unsatisfactory. In particular, it does not describe the strong suppression of the oscillations in the mixed state below  $B_{c2}$ . To demonstrate this suppression directly, Fig. 1(c) shows the result of fitting the LK theory to the experiment over the normal region only, and then extrapolating the result into the superconducting region. A comparison of Fig. 1(c) with the experiment of Fig. 1(a) shows an excellent fit to the experiment in the normal region, but there is an appreciable attenuation of the quantum oscillations in the superconducting region with respect to that in the extrapolated LK theory. A similar suppression of the signal has also recently been observed in V<sub>3</sub>Si (Ref. 17) (see discussion below). The reduction in the amplitude of the dHvA oscillations in the mixed state due to the spatial inhomogeneity of B in Eq. (1) can be shown<sup>19</sup> to be small, compared to the observed reduction, under the experimental conditions (large  $\kappa$ ,  $B \sim B_{c2}$ ). The additional attenuation can be described with a damping term<sup>17</sup> present only in the vortex state,

$$R_{S} = \exp(-\pi/\omega_{c}\tau_{S}) . \tag{3}$$

In the Lifshitz-Kosevich framework,  $R_S$  corresponds to an additional relaxation or scattering felt by the quasiparticles in the vortex state and  $\tau_S^{-1}$  is the corresponding relaxation rate. Figures 3(a)-3(c) show the results of a detailed analysis, for a number of field sweeps of the amplitude of three frequency components in Fig. 1(a) as a function of magnetic field  $B_0$ . We find that the effect of the superconductivity is to introduce an additional field-dependent relaxation or scattering of the quasiparticles. Table I shows the results of analyzing the oscillations in the vortex state with Eqs. (1), (2), and (3). Within the experimental error, there is no change in the extremal areas or effective-masses of the orbits observed on entering the

TABLE I. Measured dHvA frequencies F, associated cyclotron masses  $m^*$  and scattering rates  $\tau^{-1}$  (determined from the field and temperature dependencies of the dHvA signals), for the normal  $(B_0 = 24.1 \text{ T})$  and vortex  $(B_0 = 14.6 \text{ T})$  states. The magnetic field is oriented along [001] of the cubic (room temperature) phase.  $\Gamma X = 0.59 \text{ Å}^{-1}$ . The average Fermi velocity and wave vector are given by  $\langle v_F \rangle = (2\hbar e F)^{1/2}/m^*$  and  $\langle k_F \rangle = (A/\pi)^{1/2}$ , respectively, where  $A = (2\pi e/\hbar)F$  and  $l_0 = v_F \tau_0$ .

F (T) (normal)	F (T) (vortex)	$\langle k_F \rangle \ (\text{\AA}^{-1})$	m* (m <sub>e</sub> ) (normal)	m* (m <sub>e</sub> ) (vortex)	$ au_0^{-1}$ (THz)	$v_F$ (10 <sup>5</sup> ms <sup>-</sup>	<sup>1</sup> ) l <sub>0</sub> (nm)	$\tau_S^{-1}$ (THz) (B=15.2 T)
244±6	238±6	0.09	0.9±0.4	0.9±0.3	1.3±0.4	1.1±0.4	84±40	
344±6	338±6	0.1	$1.0 \pm 0.6$	$1.0 \pm 0.3$	$1.2 \pm 0.4$	$1.2 \pm 0.4$	100±47	$0.36 \pm 0.1$
581±10	573±10	0.13	1.10±0.06	$1.3 \pm 0.2$	$1.7 \pm 0.1$	$1.4 \pm 0.2$	82±12	$0.44 \pm 0.1$
679±10	670±12	0.14	$1.9 \pm 0.2$	$1.7 \pm 0.4$	$2.0 \pm 0.1$	$0.9 \pm 0.2$	45±10	$0.57 \pm 0.1$
809±10	798±11	0.16	$1.7 \pm 0.2$	$2.1 \pm 0.3$	$0.7 \pm 0.1$	$1.1 \pm 0.2$	157±36	$0.56 \pm 0.1$
1162±20			$2.0 \pm 0.2$	$1.3 \pm 0.8$				
1618±20			5±2					

superconducting state.

A semiclassical interpretation of our results points to the existence of coherent Landau-quantized quasiparticles in the vortex state. Since the dHvA oscillations have

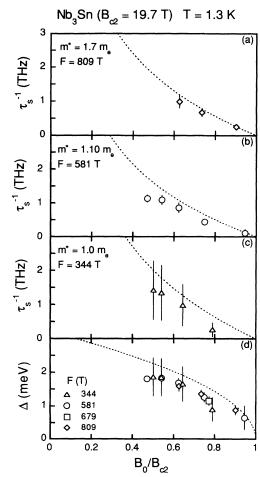


FIG. 3. (a)–(c) Variation of the additional quasiparticle damping  $\tau_S^{-1}$  with magnetic field  $B_0$  in Nb<sub>3</sub>Sn for a number of dHvA frequencies. Dotted lines are the predictions of Eqs. (4) and (5) assuming  $\Delta(0)=3.2$  meV and  $B_{c2}=19.7$  T. (d) The orbitally averaged gap parameter  $\Delta_{\rm orbit}$  determined from Eq. (4) and the experimentally measured  $R_S$ . Experimental points are the average of 30 field pulses taken at different values of  $B_{\rm max}$  and  $\partial B/\partial t$ .

the same frequencies and associated masses as those observed in the normal state, the quasiparticle states in the normal and mixed phase appear to be closely related. Under the experimental conditions ( $B_0 = 15$  T), the Landau-level separation  $\hbar\omega_c \approx 1.5$  meV, the (spatially averaged) energy gap  $\Delta(B_0 = 15 \text{ T}) \approx 1.6 \text{ meV}$  and the measured additional quasiparticle scattering  $\hbar \tau_S^{-1} \approx 0.3$ meV. Within a conventional Lifshitz-Kosevich framework, the existence of dHvA oscillations signals the presence of low-energy quasiparticle excitations. The electronic excitations in the mixed state have been investigated theoretically by a number of workers.<sup>3-7</sup> In a clean type-II material, the spatially varying order parameter in the mixed state can lead to the existence of excitations below  $\Delta$ , 6,7,21,22 another viewpoint is that excitations can result from the overlap of the bound states<sup>4</sup> associated with the vortex cores.

There has been recent theoretical progress in understanding the properties of superconductors in high magnetic fields.<sup>7</sup> The effects of Landau quantization persist in the mixed state and lead to an oscillatory contribution to the free energy and in the "quantum limit,"  $\hbar\omega_c \gg \Delta$ , where the inter-Landau level pairing can be ignored, the excitation spectrum is gapless. However, many features of the data are explained by the calculation of Brandt, Pesch, and Tewordt<sup>6</sup> which is based on Abrikosov's solution for the order parameter and an expansion of Green's functions. 6,22 Using the results of Brandt, Pesch, and Tewordt, Maki9 and Wasserman and Springford12 examined quantum oscillations in the superconducting state and suggested that the main effect of superconductivity (close to  $B_{c2}$ ) is to introduce an additional damping term into the dHvA amplitude as Eq. (3) where,

$$\tau_S^{-1} = \frac{\Delta_{\text{orbit}}^2 2\pi^{1/2} \Lambda}{\hbar v_F} , \qquad (4)$$

 $\Delta_{
m orbit}$  is the spatially averaged order parameter appropriate to a particular orbit on the Fermi surface  $\Delta_{
m orbit}^2 = \langle \, |\Delta|^2 \, \rangle_{
m orbit}, \ \Lambda = (2\hbar e B_0)^{-1/2}, \ {\rm and} \ v_F$  is the Fermi velocity, which we shall assume is given by the orbital average value  $\langle \, v_F \, \rangle = (2\hbar e F)^{1/2}/m^*$ . The dotted lines in Figs. 3(a)-3(c) show the prediction of this model assuming that  $\Delta_{
m orbit} = \Delta$ , where  $\Delta$  is described by the phenome-

nological expression,<sup>23</sup>

$$\Delta^2 = \Delta^2(0) \left[ 1 - \frac{B_0}{B_{C2}} \right] . \tag{5}$$

Within the model which leads to Eq. (4), we have a method to investigate the anisotropy of the superconducting gap over the Fermi surface. The measured values of  $\tau_S^{-1}(B/B_{c2})$  for the orbits in Figs. 3(a)-3(c) may now be reexpressed as  $\Delta_{\text{orbit}}(B/B_{c2})$ , using Eq. (4). Such an analysis is shown in Fig. 3(d). Plotting the data in this manner reveals that, for a given field, the gap values obtained from averaging over different pieces of Fermi surface are the same to within experimental error. For the extremal orbits in Table I, a single value of the order parameter can account for the damping of the dHvA effect on several different sheets of the Fermi surface. The dotted curve expresses the variation to be expected according to Eq. (5) using the literature value<sup>24</sup> of  $\Delta(0) = 3.2$ meV. Finally, we have reconstructed the dHvA effect signal to be expected using Eqs. (1)-(5), and  $\Delta(0)=3.2$ meV. The result in Fig. 1(d) closely resembles the experiment in Fig. 1(a). Many of the predictions of the above model (e.g., the similar frequencies and effective masses in the normal and mixed states) may also be present in other descriptions of the mixed state. 7-11,13 Also, the increasing magnitude of the order parameter with  $B_{c2}-B$  will lead to a suppression of the low-energy electronic excitations.

A recent study<sup>17</sup> of the sister compound V<sub>3</sub>Si revealed one dHvA frequency, in contrast to the seven frequencies reported here. The larger number of frequency components is due to the higher quality of the Nb<sub>3</sub>Sn sample used in this study. Our observation of many frequency components meant that the analysis presented here is carried out by least-squares fitting of the LK formula to the data rather than through spectral analysis as in Ref. 17. The limited quantum oscillatory data obtained in V<sub>3</sub>Si can also be described using Eqs. (1)–(5) for a single Fermi-surface sheet.

In summary, we have observed dHvA oscillations in the normal and vortex state of Nb<sub>3</sub>Sn. Quantum oscillations due to five extremal Fermi-state orbits clearly persist deep into the mixed state. Within the experimental error of our measurements, we observe no change in the period of the oscillations or the associated cyclotron masses (measured for  $T \ll T_c$ ). The primary effect of the superconductivity is to introduce an additional field-dependent scattering of the quasiparticles. Our results demonstrate that Landau quantization of the electronic states exists in the vortex state. In principle, quantum oscillatory measurements of this type will yield detailed information about the anisotropies of the superconducting state.

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