Electron-spin resonance of Eu$^{+2}$ spins in metallic EuCo$_{2-y}$As$_2$ single crystals

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The metallic compound EuCo$_{2-y}$As$_2$ orders into an antiferromagnetic helical structure below a Néel temperature $T_N = 40 - 45$ K. The effective magnetic moment $\mu_{\text{eff}}$ of the Eu spins-7/2 in the paramagnetic state above $T_N$ is enhanced by about 7% compared to the value expected for spectroscopic splitting factor $g = 2$, and the saturation moment at high fields and low temperatures $T$ is also sometimes enhanced. Here magnetic field ($H$)-swept X-band electron-spin-resonance (CW ESR) measurements of the microscopic magnetic properties of the Eu spins are reported that were carried out to identify the origin of the enhanced $\mu_{\text{eff}}$. In order to analyze the data, the complex field-dependent magnetic susceptibility components $\chi'$ and $\chi''$ are derived which allow the time-averaged microwave power absorption $P(H)$ to be computed, where $P(H)$ is found to contain an $H$ prefactor. As previously discussed [D. C. Johnston, arXiv:1808.05436v2], these theoretical predictions are especially relevant to broad Dysonian CW ESR spectra for local magnetic moments in metals, but are also applicable more broadly to CW ESR and NMR spectra. By fitting the CW ESR spectra for EuCo$_{2-y}$As$_2$ by this theory, the enhancement of $\mu_{\text{eff}}$ is inferred to arise from ferromagnetic polarization of the Co 3$d$ band electrons by the Eu spins, where the polarized band electrons form a spin cloud around the Eu spins that adiabatically follows the motion of the Eu spins. To determine the influence of the $H$ prefactor in $P(H)$ on the parameters obtained from the fits, the spectra were fitted both with and without the $H$ prefactor and the results compared. The theoretical prediction of $P(H)$ for broad Dysonian CW ESR spectra is further discussed in the Appendix along with the field-integrated behavior.

I. INTRODUCTION

The s-state ions Eu$^{+2}$ and Gd$^{+3}$ have the electronic configuration $4f^7$ with spin $S = 7/2$ and orbital angular momentum $L = 0$. Therefore spin-orbit coupling of these ions to the lattice is very weak and their spectroscopic splitting factors ($g$-factors) are usually both close to the free-electron value of 2. The paramagnetic effective moment for $g = 2$ and $S = 7/2$ is $\mu_{\text{eff}} = g\sqrt{S(S + 1)} \mu_B = 7.94 \mu_B$, where $\mu_B$ is the Bohr magneton. In most compounds containing these ions, magnetic susceptibility ($\chi$) measurements in the paramagnetic state reveal $\mu_{\text{eff}}$ values close to this value. For example, for the Eu$^{+2}$ spins in the paramagnetic state of the helical antiferromagnet EuCo$_2$P$_2$ with the body-centered-tetragonal ThCr$_2$Si$_2$ structure, to within the errors $\mu_{\text{eff}}$ is equal to the predicted value and the ordered (saturation) moment at low temperatures is also equal to the predicted value $\mu_{\text{sat}} = gS \mu_B = 7 \mu_B$/Eu atom [1].

However, in the isostructural helical antiferromagnet EuCo$_2$As$_2$, $\mu_{\text{eff}}$ was found to be about 8.5 $\mu_B$/Eu atom, corresponding to a 7.1% enhancement of the $g$ value [2]. From electronic structure calculations, this enhancement was found to be due to ferromagnetic polarization of the Co 3$d$ electrons by the Eu spins [2]. One can envision two scenarios for the implementation of this polarization. In one scenario, the enhancement could come from polarization of nearly-localized 3$d$ electrons around the Co atoms and in the second one, the enhancement could originate from polarization of itinerant Co 3$d$ band electrons in the vicinity of the Eu spins. In the latter case, one would expect microscopic measurements of the Eu $g$-factor to show an enhancement of about 7%, whereas in the former case not so much. The present Eu electron-spin resonance (CW ESR) measurements were carried out in the paramagnetic state of EuCo$_{2-y}$As$_2$ from 50 K to 180 K to obtain a microscopic measurement of the degree to which the Eu $g$-factor is enhanced, if at all. We found that the $g$-factor of the Eu spins is strongly temperature $T$ dependent, and that the $g$-factor averaged over temperature is enhanced above the free-electron value $g = 2$ for the two Crystals #2 and #3 that we measured by 5% and 7.1%, respectively. We infer that the enhanced $g$-factor as reflected by the enhanced effective moment in the magnetic susceptibility experiments arises from a ferromagnetically-polarized conduction-electron spin cloud around each Eu spin that adiabatically follows the motion of the Eu spin.

Because EuCo$_{2-y}$As$_2$ is metallic, in the present work we expected and found a Dysonian lineshape of applied magnetic field $H$-swept Eu CW ESR spectra at fixed microwave angular frequency $\omega$. One of us recently inferred [3] that an $H$ prefactor is present in the expression for the field-dependent time-averaged absorbed microwave power $P(H)$ that was not considered previously and has a significant influence on the calculated field-derivative lineshape ($A/B$ ratio) of the spectra. Therefore, in the present work we carried out a rigorous calculation of $P(H)$ and confirmed the presence of the $H$ prefactor, and the influence of this prefactor on the fitted values of the lineshape parameters was quantitatively tested. These parameters are the resonance field $H_{\text{res}}$, the Lorentzian half-width at half maximum peak

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height $\Delta H$, and the Dysonian parameter $\alpha$ which is the ratio of the dispersive to the absorptive magnetic susceptibilities that are present in the Dysonian absorptive susceptibility $\chi''_D(H)$. The fitted $\alpha$ parameter is found to strongly increase by up to a factor of $\sim 3$ when the $H$ prefactor in $\chi''_D(H)$ is omitted compared to that obtained when the prefactor is present, whereas $H_{\text{res}}$ and $\Delta H$ are only weakly affected when the $H$ prefactor is omitted. Furthermore, without this $H$ prefactor, the double integral of the field-derivative microwave power absorption spectrum versus field $H$ from $H = 0$ to $\infty$ gives a finite result and has frequently been used to obtain a value proportional to the dc $\chi$ in the paramagnetic state. However, when the $H$ prefactor is included as dictated by theory, the double integral is logarithmically divergent versus $H$ [3] and such analyses are problematic.

In Sec. II the experimental details are given. The well-known theory for the CW ESR power absorption versus $\omega$ at fixed $H$ obtained from $\chi''(\omega)$ at fixed $H$ is reviewed in Sec. III A. In Sec. III B we present our calculation of the CW ESR spectra obtained by scanning $H$ at fixed $\omega$ that is needed to analyze our CW ESR spectra. This theory is derived from the Bloch equations, consistent with the derivation from the Bloch equations of the theory for CW ESR carried out while scanning $\omega$ at constant $H$. An overview of the theoretically-predicted Dysonian CW ESR spectra is given in Sec. IV A. The results of our CW ESR measurements and analyses are given in the remainder of Sec. IV, including the results and analyses yielding the fitted values of $\alpha$, $H_{\text{res}}$, $g$-factor, and $\Delta H$ versus temperature $T$. A summary and discussion is given in Sec. V.

In the Appendix the theoretically-predicted $P(H)$ of Dysonian CW ESR resonance spectra is elaborated along with the field-integrated power absorption $P_{\text{int}}(H)$ from $H = 0$ to a maximum field $H_{\text{max}}$, where $P_{\text{int}}(H_{\text{max}} \to \infty)$ is finite if the theoretically-required $H$ prefactor in $P(H)$ is omitted, whereas $P_{\text{int}}(H_{\text{max}} \to \infty)$ diverges logarithmically if this prefactor is included.

## II. EXPERIMENTAL DETAILS

Two single crystals of EuCo$_{2-y}$As$_2$ labeled Crystals #2 and #3 were studied that were taken from the same batches of crystals from which extensive crystallographic and physical-property data for crystals also labeled Crystals #2 (Sn-flux-grown) and #3 (CoAs self-flux-grown) were presented in Ref. [2].

The CW ESR measurements were carried out at a fixed X-band frequency of 9.380 GHz and magnetic field scan range 0 to 6 kOe using an Elexsys E580 FT/CW EPR spectrometer in CW mode. The static magnetic field $H$ was applied along the $c$ axis of the two crystals measured. The microwave magnetic field was perpendicular to $H$ and hence directed along the two large flat surfaces of the crystals parallel to the $ab$ plane. The ESR data reported here cover the temperature $T$ range from 50 K to 180 K which is in the paramagnetic temperature region of the two crystals above their respective Néel temperatures of 45 K for Crystal #2 and 40 K for Crystal #3 [2]. To improve the signal-to-noise ratio, the applied magnetic field was modulated at a frequency of 100 kHz with lock-in amplifier detection at that frequency, so a measured ESR spectrum is the field derivative of the microwave power absorption spectrum.

In this paper we use Gaussian cgs units for magnetic properties and SI units for electrical properties.

## III. THEORY

### A. CW ESR with Variable Frequency at Fixed Field

In this section CW ESR theory is reviewed for the well-documented case of slowly sweeping the microwave magnetic field angular frequency $\omega$ (slow passage) at a fixed static uniform magnetic field

$$H_0 = H_0 \hat{k},$$

(1a)

where the unit vectors that point towards the positive Cartesian $x$, $y$, and $z$ axes are denoted as $\hat{i}$, $\hat{j}$ and $\hat{k}$, respectively. We take the microwave magnetic field to be given by

$$H_1 = H_1 \cos(\omega t) \hat{i},$$

(1b)

which is perpendicular to $H_0$ and can be considered to be a superposition of a microwave field with amplitude $H_1/2$ rotating with angular velocity $\vec{\omega} = \omega \hat{k}$ and a counter-rotating field with the same amplitude and $\vec{\omega} = -\omega \hat{k}$. In this paper we only consider small values of the amplitude $H_1$ (nonsaturating condition).

The absorptive imaginary part $\chi''$ of the complex magnetic susceptibility of the system

$$\chi = \chi' - i \chi'',$$

(2)

where $\chi'$ is the real dispersive part, is [4–8]

$$\chi''(\omega) = \frac{\gamma M_0 \Delta \omega}{(\omega - \omega_{\text{res}})^2 + \Delta \omega^2}. $$

(3a)

Here $\omega_{\text{res}} = \frac{\gamma H_{\text{int}}}{M_0}$ is the resonant angular frequency, $H_{\text{int}}$ is the magnitude of the magnetic field internal to the sample which may be different from $H_0$ due to an internal demagnetization field as discussed later in Sec. IV C, $\gamma$ is the gyromagnetic ratio for a local moment in the sample, $M_0(H_0) = \chi_0 H_0$ is the fixed static uniform volume magnetization (magnetic moment per unit volume) at the fixed field $H_0$, and $\chi_0 \equiv \chi_0(T)$ is the temperature $T$-dependent dimensionless static uniform volume magnetic susceptibility. As
the driving angular frequency \( \omega \) increases from some initial small value, the system goes through resonance at \( \omega = \omega_{\text{res}} \).

The time-averaged power absorbed by a sample versus \( \omega \) at fixed \( H_0 \) is given by [4, 6]

\[
P(\omega) = \frac{H_0^2 V}{2} \omega \chi''(\omega),
\]

where \( H_0^2 \) has dimensions of energy per unit volume and \( V \) is the volume of the sample exposed to the microwave magnetic field. This \( V \) is not necessarily the same as the volume of the sample because in electrically-conducting samples \( V \) can be limited by the skin depth \( \delta \) (see Sec. IV B below). The integrated power \( P_{\text{int}}(\omega) \) absorbed by the sample from \( \omega = 0 \) up to frequency \( \omega \) is the integral of Eq. (4) with respect to \( \omega \) given by

\[
P_{\text{int}}(\omega) = \frac{H_0^2 V}{2} \int_0^\omega \omega' \chi''(\omega')d\omega'.
\]

Inserting \( \chi''(\omega) \) from Eq. (3a) into this expression and carrying out the integration gives

\[
P_{\text{int}}(\omega) = \frac{\gamma M_0 H_0^2 V}{2} \omega_{\text{res}} \left\{ \arctan \left( \frac{\omega - \omega_{\text{res}}}{\Delta \omega} \right) + \arctan \left( \frac{\omega_{\text{res}}}{\Delta \omega} \right) \right\} + \frac{\Delta \omega}{2} \ln \left[ \frac{\Delta \omega^2 + (\omega - \omega_{\text{res}})^2}{\Delta \omega^2 + \omega_{\text{res}}^2} \right].
\]

A plot of the quantity in curly brackets in Eq. (6) versus \( \omega \) for \( \omega = 0 \) to 20 units is shown in Fig. 1(a) for \( \omega_{\text{res}} = 1 \) unit and \( \Delta \omega = 1 \) unit.

For \( \omega \gg \omega_{\text{res}} \) and \( \omega \gg \Delta \omega \), Eq. (3a) gives

\[
\omega \chi''(\omega) \sim \frac{\Delta \omega}{\omega}.
\]

Thus at high frequencies, \( P_{\text{int}}(\omega) \) exhibits a logarithmic divergence described by

\[
P_{\text{int}}(\omega) \sim \Delta \omega \ln \omega
\]

and a plot of \( \exp[P_{\text{int}}(\omega)] \) versus \( \omega \) should become linear in \( \omega \) for \( \omega \gg \omega_{\text{res}} \) and \( \omega \gg \Delta \omega \), as verified in Fig. 1(b).

If the variable \( \omega \) prefactor in Eq. (4) were instead set to the fixed resonant frequency \( \omega_{\text{res}} \), Eq. (5) would give

\[
P_{\text{int}}(\omega) = \frac{\gamma M_0 H_0^2 V}{2} \omega_{\text{res}} \left[ \arctan \left( \frac{\omega - \omega_{\text{res}}}{\Delta \omega} \right) + \arctan \left( \frac{\omega_{\text{res}}}{\Delta \omega} \right) \right],
\]

which yields the finite limiting value

\[
P_{\text{int}}(\omega \rightarrow \infty) = \frac{\gamma M_0 H_0^2 V}{2} \omega_{\text{res}} \left[ \frac{\pi}{2} + \arctan \left( \frac{\omega_{\text{res}}}{\Delta \omega} \right) \right],
\]

instead of the correct logarithmic divergence in Eq. (8).

### B. CW ESR with Variable Field at Fixed Frequency

In a CW ESR spectrometer such as used in our experiments, one scans the applied magnetic field \( H_0 \) at fixed \( \omega \) instead of scanning \( \omega \) at fixed \( H_0 \). Physically, one expects the same behaviors for the time-averaged microwave power absorption \( P(H_0) \) and the field-integrated microwave power absorption \( P_{\text{int}}(H_0) \) at fixed frequency as for \( P(\omega) \) and \( P_{\text{int}}(\omega) \) at fixed \( H_0 \) above and in the preceding section. Here the dispersive \( \chi'(H_0) \) and absorptive \( \chi''(H_0) \) components of the complex magnetic-field-dependent magnetic susceptibility in Eq. (2) at fixed \( \omega \) are derived following the same steps as utilized in Ref. [6] to derive \( \chi(\omega) \) at fixed \( H_0 \), and from those the time-averaged microwave power \( P(H_0) \) absorbed by the sample at fixed \( \omega \) is calculated.

The Bloch equations describe the motion of the volume magnetization vector \( \mathbf{M} \) under the influence of a total applied magnetic field \( \mathbf{H} \) in the presence of longitudinal (z-axis, parallel to \( \mathbf{H} \)) \((1/T_1)\) and transverse (xy plane, perpendicular to \( \mathbf{H} \)) spin-spin \((1/T_2)\) relaxation rates. The Cartesian components of \( \mathbf{M} \) are given by the Bloch equations as [4–6]

\[
\frac{dM_x}{dt} = \gamma (\mathbf{H} \times \mathbf{M})_z - \frac{M_x}{T_2},
\]

\[
\frac{dM_y}{dt} = \gamma (\mathbf{H} \times \mathbf{M})_y - \frac{M_y}{T_2},
\]

\[
\frac{dM_z}{dt} = \gamma (\mathbf{H} \times \mathbf{M})_z + \frac{M_0 - M_z}{T_1}.
\]

Here,

\[
\mathbf{H} = \mathbf{H}_0 + \mathbf{H}_1,
\]

where the applied and microwave magnetic fields \( \mathbf{H}_0 \) and \( \mathbf{H}_1 \), respectively, are given in Eqs. (1). According to Fig. 2, the fixed angular frequency \( \omega \) is the resonant frequency \( \omega_{\text{res}} \). The corresponding magnitude of the applied magnetic field at resonance is

\[
\omega_{\text{res}} = \omega_{\text{res}}/\gamma.
\]

Here \( \omega_{\text{res}} \) includes the contribution of the macroscopic internal demagnetization field.
FIG. 1: (a) Frequency-integrated microwave power absorption $P_{\text{int}}(\omega)$ [the quantity in curly brackets in Eq. (6)] up to frequency $\omega$ versus $\omega$ for $\omega_{\text{res}} = 1$ unit and $\Delta \omega = 1$ unit. (b) $\exp[P_{\text{int}}]$ versus $\omega$ plotted using the data in (a). The high-$\omega$ data are seen to approach linearity in $\omega$ as predicted by Eq. (8).

FIG. 2: Relationship of the fixed microwave quantum $E_\omega = \hbar \omega = \hbar \omega_{\text{res}}$ to the resonant field $H_{\text{res}} = \omega_{\text{res}}/\gamma$ in a Zeeman energy diagram for an illustrative spin $S = 1/2$. The ESR intensity decreases as the variable applied dc field $H$ deviates from $H_{\text{res}}$, because then the required optimal power absorption relation $H = H_{\text{res}}$ is no longer fulfilled.

In a primed frame of reference rotating about the $z'$ axis at fixed angular velocity $\omega_{\text{res}} = \omega_{\text{res}} k = \omega_{\text{res}} k'$, the so-called effective magnetic field $H_{\text{eff}}$ is

$$H_{\text{eff}} = H_1 \hat{z}' + \left( H_0 - \frac{\omega_{\text{res}}}{\gamma} \right) \hat{k}'$$  \hspace{1cm} (13)$$

The Bloch equations (10) in this reference frame become

$$\frac{dM_{x'}}{dt} = -\gamma (H_0 - H_{\text{res}}) M_{y'} - M_{x'} \frac{T_2}{T_2},$$

$$\frac{dM_{y'}}{dt} = \gamma (H_0 - H_{\text{res}}) M_{x'} - \gamma H_1 M_{z'} - M_{y'} \frac{T_2}{T_2},$$

$$\frac{dM_{z'}}{dt} = \gamma H_1 M_{y'} + \frac{M_0 - M_{z'}}{T_1}.$$ \hspace{1cm} (14)

These equations are solved under slow-passage conditions. Substituting $H_{\text{eff}}$ from Eq. (13) into Eqs. (14), one obtains

$$0 = -\gamma (H_0 - H_{\text{res}}) M_{y'} - M_{x'} \frac{T_2}{T_2},$$

$$0 = \gamma (H_0 - H_{\text{res}}) M_{x'} - \gamma H_1 M_{z'} - M_{y'} \frac{T_2}{T_2},$$

$$0 = \gamma H_1 M_{y'} + \frac{M_0 - M_{z'}}{T_1}.$$ \hspace{1cm} (16)

Solving these three simultaneous equations for the Cartesian components of $\mathbf{M'}$ and retaining only terms up to first order in $H_1$ (nonsaturating condition) gives the steady-state solutions in the rotating frame of reference as

$$M_{x'} = M_0 H_1 \frac{(H_0 - H_{\text{res}})(\gamma T_2)^2}{(H_0 - H_{\text{res}})^2(\gamma T_2)^2 + 1},$$

$$M_{y'} = -M_0 H_1 \frac{\gamma T_2}{(H_0 - H_{\text{res}})^2(\gamma T_2)^2 + 1},$$

$$M_{z'} = M_0.$$ \hspace{1cm} (17)

The half-width of the Lorentzian resonance line at half-height when $H_0$ is varied and $\omega = \omega_{\text{res}}$ is fixed is

$$\Delta H_0 = \frac{1}{\gamma T_2}$$ \hspace{1cm} (18a)$$

and the volume magnetization $M_0$ is given by

$$M_0 = \chi_0 H_0.$$ \hspace{1cm} (18b)
where as noted above $\chi_0 \equiv \chi_0(T)$ is the dimensionless $T$-dependent magnetic susceptibility per unit volume. Using Eqs. (18), Eqs. (17) become

$$M_x'(H_0) = \chi_0 H_1 H_0 \frac{H_0 - H_{\text{res}}}{(H_0 - H_{\text{res}})^2 + \Delta H_0^2},$$
$$M_y'(H_0) = -\chi_0 H_1 H_0 \frac{\Delta H_0}{(H_0 - H_{\text{res}})^2 + \Delta H_0^2},$$
$$M_z'(H_0) = \chi_0 H_0.$$

The dispersive $\chi'(H_0)$ and absorptive $\chi''(H_0)$ parts of the complex magnetic susceptibility in Eq. (2) are obtained as

$$\chi'(H_0) = M_x'(H_0)/H_1,$$
$$\chi''(H_0) = -M_y'(H_0)/H_1.$$

Then Eqs. (19) give

$$\chi'(H_0) = \chi_0 H_0 \frac{H_0 - H_{\text{res}}}{(H_0 - H_{\text{res}})^2 + \Delta H_0^2},$$
$$\chi''(H_0) = \chi_0 |H_0| \frac{\Delta H_0}{(H_0 - H_{\text{res}})^2 + \Delta H_0^2},$$

where the prefactor $H_0$ in Eq. (21b) was modified to $|H_0|$ in order for $\chi''(H_0)$ to be even in $H_0$ as required physically.

Note that the variable field $|H_0|$ is contained as a prefactor within the expression for the absorptive susceptibility $\chi''(H_0)$ in Eq. (21b). However, when $\omega$ is variable and $H_0$ is held constant as discussed above in Sec. III A, the $\omega$ prefactor does not appear as a prefactor in $\chi''(\omega)$ in Eq. (3a), but instead appears as a prefactor in the combination $\omega \chi''(\omega)$ in Eq. (4) for the microwave power $P(\omega)$ absorbed by the sample.

One sees that the prefactors in $\chi''(\omega)$ with fixed $H_0$ in Eq. (3a) and $\chi''(H_0)$ at fixed $\omega$ in Eq. (21b) are the same. The fraction in $\chi''(\omega)$ at fixed $H_0$ can be derived from the fraction in $\chi''(H_0)$ at fixed $\omega$ by making the replacements $H_0 \rightarrow \omega/\gamma$, $H_{\text{res}} \rightarrow \omega_{\text{res}}/\gamma$, and $\Delta H_0 \rightarrow \Delta \omega/\gamma$, and conversely.

The steady-state $x$-axis magnetization component in the laboratory reference frame is [4]

$$M_x(H_0) = H_1 \left[ \chi'(H_0) \cos(\omega_{\text{res}} t) + \chi''(H_0) \sin(\omega_{\text{res}} t) \right].$$

From Eqs. (1) with $\omega \rightarrow \omega_{\text{res}}$ and Eq. (11), one obtains

$$\frac{dH}{dt} = \frac{dH_{1z}}{dt} = -H_1 \omega_{\text{res}} \sin(\omega_{\text{res}} t).$$

The time $t$-dependent microwave power absorbed by a sample is calculated from [4, 6]

$$P(H_0, t) = -V M(H_0) \frac{dH(t)}{dt} = -V M_x(H_0) \frac{dH_{1z}}{dt}.$$  (24)

Using Eqs. (22) and (23), taking the time average of Eq. (24) gives the time-averaged microwave power absorbed by the sample as

$$P(H_0) = \frac{H_1^2 V \omega_{\text{res}}}{2} \chi''(H_0),$$  (25)

where $\chi''(H_0)$ is given above in Eq. (21b). This shows that $P(H_0)$ is proportional to $\chi''(H_0)$, which is the first term in Eq. (26) below with definition $H_0 \rightarrow H$ and $\alpha = 0$.

If one sets the variable $|H| = H$ prefactor in Eq. (26) below to the fixed value $H = \omega_{\text{res}}/\gamma = H_{\text{res}}$, many of the $\alpha$ values in Fig. 4 below obtained from fitting our ESR spectra without the $H$ prefactor in $\chi''(H)$ are invalid, and the expression for the field-integrated microwave power absorption (see Appendix) is also invalid.

IV. RESULTS AND ANALYSES

A. ESR Spectra Overview

The field-derivative spectra for the two crystals #2 and #3 of EuCo$_2$-yAs$_2$, each at $T = 50$ K and 180 K, are shown in Fig. 3 and exhibit Dysonian lineshapes. The Dysonian absorptive susceptibility $\chi''_D(H)$ including both the rotating and counter-rotating components of the linearly-polarized microwave field $H_1$ applied perpendicular to the applied field direction using Eq. (21b) and $H_0 \rightarrow H$ is [3, 9, 10]

$$\chi''_D(H) = \chi_0 |H| \left[ \frac{\Delta H - \alpha(H_{\text{res}} - H)}{\Delta H^2 + (H_{\text{res}} - H)^2} \right. + \frac{\Delta H - \alpha(H_{\text{res}} + H)}{\Delta H^2 + (H_{\text{res}} + H)^2}.$$  (26)

The Dysonian lineshape parameter is $\alpha = 0$ if the skin depth $\delta$ is much larger than the sample dimension perpendicular to the microwave magnetic field, and $\alpha = 1$ if $\delta$ is much smaller than this sample dimension. Here, $\chi_0 \equiv M_0/H$ is again the $T$-dependent dimensionless volume magnetic susceptibility of the sample that can be field dependent and $M_0$ is the magnitude of the thermal-equilibrium volume magnetization in the direction of the applied magnetic field in the absence of the microwave field. For the temperatures and fields of the present ESR experiments, $\chi_0$ is independent of $H$ [2]. In general, the parameters $\chi_0$, $\alpha$, $\Delta H$, and $H_{\text{res}}$ are all temperature dependent.

The parametric equation for the path of the magnetization vector in undamped ESR is given and illustrated in Ref. [11]. When damping is included, the time-average microwave power $P(H)$ absorbed in steady state by a metallic sample containing a large concentration of local magnetic moments is obtained from Eq. (25) as

$$P(H) = \frac{H_1^2 (A \delta) \omega_{\text{res}}}{2} \chi''_D(H).$$  (27)

where $V = A \delta$, $A$ is the area of a surface of the sample parallel to the linearly-polarized applied microwave field $H_1$, $\delta$ is the $T$-dependent skin depth at the microwave frequency of $H_1$ discussed in the following section, and
FIG. 3: Eu$^{+2}$ field-derivative ESR spectra (open circles) at temperatures of 50 K and 180 K for (a) a Sn-flux-grown crystal #2 and (b) a CoAs self-flux-grown crystal #3 of EuCo$_2$−$_y$As$_2$. The fits by Eq. (28) are shown as solid black curves.

FIG. 4: Ratio $\alpha$ of the dispersive susceptibility $\chi'$ to the absorptive susceptibility $\chi''$ in the Dysonian absorptive susceptibility $\chi''_D(H)$ in Eq. (26) for Crystals #2 and #3 versus temperature $T$ both with and without the $H$ prefactor in $\chi''_D(H)$. All data above $\alpha = 1$ are explicitly invalid. The correct values are the filled symbols obtained by fitting the field-derivative spectra by Eq. (28) including the $H$ prefactor.

hence $A\delta$ is the volume $V$ of the sample beneath the surface $A$ that is exposed to the microwave field. If the sample is in the shape of a rectangular prism with $H_1$ parallel to two of the faces as in the present ESR experiments, the area $A$ would by multiplied by two as long as $\delta$ is less than half the thickness of the sample perpendicular to $H_1$. The microwave power absorption obtained using the Dysonian absorptive susceptibility is discussed in more detail in the Appendix.

Our experimental field-derivative microwave power-absorption spectra at each fixed temperature for $H > 0$ were fitted by the field-derivative of the Dysonian lineshape function in Eq. (26), i.e.,

$$\frac{dP}{dH} = a + b \frac{d}{dH} \left\{ H \left[ \frac{\Delta H - \alpha(H_{\text{res}} - H)}{\Delta H^2 + (H_{\text{res}} - H)^2} \right] \right.$$

$$\left. + \frac{\Delta H - \alpha(H_{\text{res}} + H)}{\Delta H^2 + (H_{\text{res}} + H)^2} \right\}$$

(28)

where $a$ is the instrumental zero-signal offset and $b$ is the amplitude of the $dP/dH$ spectrum. The other fitting parameters at each temperature are $\Delta H$, $\alpha$ and $H_{\text{res}}$. Examples of the fits are shown for crystals #2 and #3, each at $T = 50$ K and 180 K, in Figs. 3(a) and 3(b) as solid black curves, respectively. For both crystals, the fits are very good, except for an additional feature of unknown origin for Crystal #3 in Fig. 3(b) at $H \sim 1–2$ kOe that disappears above $\sim 70$ K. As shown in Fig. 5 below, when this signal disappears the resonance field and the $g$-factor both exhibit discontinuities at $\approx 65$ K in their temperature dependences. Thus we infer that the signal at 1–2 kOe below 65 K in Fig. 3(b) for Crystal #3 does not arise from paramagnetic impurities, but is more likely associated with a phase transition at $\approx 65$ K in this crystal.

From Fig. 3, one sees that the linewidth increases with increasing $T$ for each of the two crystals. Indeed, the resonances for the two crystals at $T = 180$ K are cut off due to the $H = 6$ kOe upper limit of our measurements. From the different ordinate scales for the two temperatures in each of Figs. 3(a) and 3(b), the signal amplitude strongly decreases with increasing temperature for each crystal. Such a decrease is expected from the Curie-Weiss $T$ dependence [2] of the Eu$^{+2}$ spin susceptibility $\chi_0$ in Eq. (26).

B. $\alpha$ Parameter

Shown in Fig. 4 are plots of $\alpha$ versus $T$ obtained from fits to the field derivative of the microwave absorption
spectra such as in Fig. 3 using Eq. (28) for each of Crystals #2 and #3 both with and without the $H$ prefactor in Eq. (28). The values obtained without the $H$ prefactor are a factor of two to three large compared to the correct values obtained with the $H$ prefactor included in the fits. Furthermore, the value of $\alpha$ must be in the range $0 \leq \alpha \leq 1$ for a physically-valid fit, so the values of $\alpha > 1$ obtained without the prefactor in Fig. 4 are explicitly invalid.

The skin depth $\delta(H, T)$ in Eq. (27) is given in SI units by

$$\delta(H, T) = \sqrt{\frac{\rho(T)}{\pi f \mu(H, T)}},$$

(29)

where $\rho$ is the electrical resistivity, $f$ is the microwave frequency, and $\mu$ is the magnetic permeability of the sample. The magnetic permeability in SI units is given by

$$\mu = \mu_0 [1 + (M/H)],$$

(30)

where $\mu_0$ is the magnetic permeability of free space. $M$ is the volume magnetization of the sample and in general the dimensionless ratio $M(T, H)/H$ can be dependent on the temperature $T$ and the magnitude and direction of the applied field $H$. The required SI value of $M/H$ is obtained from $M/H$ in dimensionless cgs units via $M/H (\text{SI}) = (4\pi)^{-1}(M/H)$ (cgs).

For EuCo$_2$-yAs$_2$ in the field and paramagnetic temperature ranges of interest in this paper, $M/H$ in Eq. (30) is just the magnetic susceptibility per unit volume $\chi_V$ that is given by the Curie-Weiss law in cgs units as

$$\chi_V = \frac{C}{V_M (T - \theta)}.$$  

(31a)

$$C \approx 9.0 \text{ cm}^3 \text{ K} \text{ mol Eu}^{-1},$$

(31b)

$$\theta \approx 22 \text{ K},$$

(31c)

$$V_M \approx 52.6 \text{ cm}^3 \text{ mol Eu}^{-1},$$

(31d)

where the approximate values of the molar Curie constant $C$ and the Weiss temperature $\theta$ averaged over data for five crystals and over the two field directions $H \parallel c$ and $H \parallel ab$ and the molar volume $V_M$, all from Ref. [2], are given in the last three of Eqs. (31). Then Eq. (31a) gives

$$\chi_V = \frac{0.17 \text{ K}}{T - 22 \text{ K}}.$$  

(32)

At a temperature of 50 K in the paramagnetic state where $\chi_V$ is near its maximum value versus temperature, the value of $\chi_V$ is

$$\chi_V (50 \text{ K}) = 0.0060 \text{ (cgs)}.$$  

(33)

so the $M/H$ term in Eq. (30) can be set to zero. Inserting our X-band microwave frequency $f = 9.390$ GHz and the value for $\mu_0$ into Eq. (29), one obtains

$$\delta(T)[\mu m] = 0.5194 \sqrt{\rho(T)[\mu\Omega \text{ cm}].}$$

(34)

When the static field $H$ is applied along the $c$ axis as in this paper, the microwave magnetic field $H_1$ is parallel to the $ab$ plane. Since the Poynting vector associated with the skin depth is normal to a surface that is perpendicular to $H$, the microwave electric field associated with $H_1$ is also oriented in the $ab$ plane. Hence the relevant resistivity in Eq. (34) for $H \parallel c$-axis is the in-plane electrical resistivity $\rho_{ab}$, which was measured for two crystals with similar results [2] that together are approximated for the temperature range $50 \text{ K} \leq T \leq 300 \text{ K}$ by the linear relation

$$\rho_{ab}(T) \approx 14 \mu \text{\Omega cm} + \left(0.083\frac{\mu\Omega \text{ cm}^2}{\text{K}}\right) T.$$  

(35)

Using Eqs. (34) and (35), one obtains $\delta = 2.2 \mu m$ and $3.2 \mu m$ at 50 K and 300 K, respectively. These values are much smaller than the $ab$-plane dimensions of our crystals ($\sim \text{mm}$) which would therefore predict $\alpha = 1$. The reason we obtain $\alpha \sim 0.4$--0.5 in Fig. 4 is not clear.

C. Resonance Magnetic Field and $g$ Factor

The resonance magnetic field $H_{\text{res}}$ versus temperature obtained from fits of the field-derivative ESR spectra by Eq. (28) is plotted for Crystals #2 and #3 in Fig. 5(a) determined both with and without the $H$ prefactor in Eq. (28). The data show clear evidence for a second-order phase transition in Crystal #3 at $\approx 65 \text{ K}$ that is not present in the data for Crystal #2. Omission of the $H$ prefactor gives an error of only $\leq 2\%$ in the fitted $H_{\text{res}}$ as shown. This error propagates to a similar error after the demagnetizing field is corrected for as follows.

Due to the presence of highly magnetic Eu$^{+2}$ ions with spin $S = \frac{7}{2}$, the magnetic field $H_{\text{int}}$ internal to a sample can be significantly different from the applied field $H$. The components of $H_{\text{int}}$ along the principal-axes directions $\beta$ are given in Gaussian cgs units by [12]

$$H_{\text{int}}^\beta = H_\beta - 4\pi N_{d\beta}M_\beta,$$

(36)

where the $\beta$ principal-axis magnetization component $M_\beta$ is in cgs units of Gauss and the magnetometric demagnetization factor $N_{d\beta}$ is in SI units where $0 \leq N_{d\beta} \leq 1$ and $\sum_{\beta=1}^3 N_{d\beta} = 1$. The dimensions and c-axis $N_{d\beta}$ values of the two crystals discussed in this paper where $H \parallel c$ axis were calculated using the expression derived in Ref. [13] and are listed in Table I.

At the resonance field $H_{\text{res}}$, Eq. (32) gives the magnetization per unit volume $M_c$ as

$$M_c = \chi_V H_{\text{res}} = \frac{(0.17 \text{ K})H_{\text{res}}}{T - 22 \text{ K}}.$$  

(37)

For example, at $T = 50 \text{ K}$, using Eq. (37) and taking $H_{\text{res}} = 3.25 \text{ kOe}$ from Fig. 5(a) and $N_{d\beta} \approx 0.75$ from Table I, one obtains $4\pi N_{d\beta}M_c \approx 0.19 \text{ kOe}$ for this term in Eq. (36), which is a large fraction of the temperature-induced change in $H_{\text{res}}$ in Fig. 5(a). Thus taking the
FIG. 5: (a) The measured Eu$^{+2}$ resonance field $H_{\text{res}}$ versus temperature $T$ for Crystals #2 (red circles) and #3 (blue squares) of EuCo$_{2-x}$As$_2$ obtained from fits of the field-derivative spectra by Eq. (28) both with (filled symbols) and without (open symbols) the $H$ prefactor. (b) The internal resonance field $H_{\text{int}}$ obtained from the data including the $H$ prefactor in panel (a) after correction of the demagnetizing field according to Eq. (36). (c) The Eu$^{+2}$ $g$-factors for Crystals #2 and #3 versus $T$ calculated from the data in panel (b) using Eq. (38b).

demagnetizing field into account results in a significant correction to the measured $H_{\text{res}}(T)$ for our crystals.

Shown in Fig. 5(b) are data for the internal resonance field $H_{\text{int}}$ versus temperature obtained from the data in Fig. 5(a) derived using Eqs. (36) and Eq. (37) when the $H$ prefactor is included in Eq. (28). One sees an unusual nonmonotonic variation of $H_{\text{int}}$ with temperature.

The $g$-factor of the Eu$^{+2}$ spins is obtained from the quantum condition

$$g = \frac{hf}{\mu_B H_{\text{int}}},$$

(38a)

where $h$ is Planck’s constant, $f$ is the microwave frequency, and $\mu_B$ is the Bohr magneton. In our experiments, the X-band microwave frequency was $f = 9.390$ GHz. Thus one obtains

$$g = \frac{6.709}{H_{\text{int}} \text{(kOe)}}.$$  

(38b)

Plots of the $g$-factor versus $T$ for Crystals #2 and #3 obtained from the data in Fig. 5(b) using Eq. (38b) are shown in Fig. 5(c), where the nonmonotonic variation in Fig. 5(b) is inverted. A discontinuity in the slope of $g$ versus temperature is seen for Crystal #3 at $T \approx 65$ K that is not present for Crystal #2, again suggesting a second-order phase transition in Crystal #3 as indicated from the resonance field data in Figs. 5(a) and 5(b). One sees $g$ values enhanced above the expected $g \approx 2$ at both low and high temperatures, with temperature-averaged values $g_{\text{ave}} = 2.10$ and 2.15 for Crystals 2 and 3, respectively. These 5.0–7.5% enhancements above the value $g = 2$ are consistent with the $\approx 7.1\%$ enhancements of the Eu$^{+2}$ effective moment in EuCo$_{2-y}$As$_2$ crystals obtained [2] from magnetic susceptibility measurements that was theoretically attributed to spin polarization of the Co 3$d$-band electrons by the Eu spins [2]. This agreement indicates that the enhancement of the Eu$^{+2}$ effective moment arises from a conduction-electron polarization cloud that surrounds each Eu moment and is ferromagnetically aligned with it, and that the Eu moment and polarization cloud respond as a single composite moment to an applied magnetic field in the paramagnetic state. If the polarized $d$ electrons instead resided elsewhere, e.g. near the Co atoms, the observed enhanced effective moment would presumably not produce such a large enhancement of the Eu-moment $g$-factor.

| Crystal | $ab$-plane | $c$-axis | $N_{\text{dc}}$ |
|---------|------------|----------|--------------|
| #2 EuCo$_{1.99(2)}$As$_2$ | $1.6 \times 6.7$ | 0.47 | 0.71 |
| #3 EuCo$_{1.92(4)}$As$_2$ | $2.5 \times 4.4$ | 0.35 | 0.79 |

*a* Grown in Sn flux with H$_2$-treated Co powder

*b* Grown in CoAs flux with H$_2$-treated Co powder
D. Linewidth

The Lorentzian half-width of the resonance $\Delta H$ versus $T$ is plotted in Fig. 6(a) for Crystals #2 and #3 obtained from fitting the ESR spectra by Eq. (28), each both with and without the $H$ prefactor in Eq. (28). Little difference is observed between the values determined with and without the $H$ prefactor in Eq. (28). The data corrected for the demagnetization field as in Fig. 5(b) are shown in Fig. 6(b). The latter data for Crystal #2 suggest possible phase transitions at $\approx 90$ K and 120 K, whereas the data for Crystal #3 do not exhibit clear evidence for any phase transitions. Overall, the linewidth of both crystals increases with increasing temperature above 70 K as expected for relaxation of the Eu spins by exchange interactions with the conduction electrons. However the behavior is not linear in $T$ as expected for such Korringa relaxation. The average slope between 90 K and 180 K for Crystal #2 is 8.5 Oe/K, whereas for Crystal #3 the average slope is 6.6 Oe/K. These slope values are in the range found for similar Fe-based ThCr$_2$Si$_2$-structure pnictide compounds containing Eu$^{+2}$ ions [14, 15]. The ratio $\Delta H^\text{int}/H^\text{int}_\text{res}$ is plotted versus $T$ in Fig. 6(c) for the two crystals. This ratio increases from about 0.25 at $T \approx 60$ K to about 0.6 at $T = 180$ K.

V. SUMMARY

The theory for CW ESR under the condition of varying microwave frequency $\omega$ at fixed static applied field $H$ is well known. This theory was reviewed in Sec. III A, where we showed that the frequency-integrated time-averaged power absorption $P_{\text{int}}(\omega)$ diverges logarithmically as $\omega$ increases beyond the resonance frequency $\omega_{\text{res}}$, contrary to what might have been expected. This divergence is due to the presence of an $\omega$ prefactor in the expression for the time-averaged power $P(\omega)$. One of us pointed out in Ref. [3] that under the experimental conditions where CW ESR spectra are obtained by scanning $H$ at constant $\omega$, there is an $H$ prefactor in the expression for $P(H)$ that causes $P_{\text{int}}(H)$ to diverge logarithmically with increasing $H$, in analogy to the former case where $\omega$ was varied at constant $H$. Furthermore, it was shown that the presence of the $H$ prefactor drastically changed the lineshape for broad ESR spectra for local moments in metals. In Sec. III B the detailed theory for the latter case was presented that rigorously confirmed the analysis in Ref. [3] and placed it on a firm footing. We then utilized the results in Sec. III B to analyze the experimental CW ESR data obtained by varying $H$ at constant $\omega$ presented for EuCo$_{2-y}$As$_2$ in subsequent sections.

Excellent fits of the experimental field-derivative CW ESR spectra for EuCo$_{2-y}$As$_2$ by the general Eq. (28) were obtained in the paramagnetic phase at temperatures from 50 K to 180 K for Crystals #2 and #3. This equation, as discussed in Ref. [3] and here in Sec. III B, contains the above-noted theoretically-required magnetic-
field $H$ prefactor omitted previously to our knowledge when fitting broad ESR spectra for local moments in metals. The influence of this $H$ prefactor on the parameters obtained from the fits of the ESR spectra for EuCo$_{2-y}$As$_2$ was tested. We find that whether or not this prefactor is included in the fit function gives only a few percent difference at most in the fitted values of $\Delta H$ and $H_{\text{res}}$, but the fitted value of $\alpha$ without the $H$ prefactor is up to a factor of three too large compared to the correct values obtained when the prefactor is included in the fit function. Some of the fitted $\alpha$ values obtained in the absence of the $H$ prefactor exceed the physical upper limit of unity for $\alpha$. An important theoretical result is that the field-integrated microwave absorption diverges logarithmically with increasing field when the required $H$ prefactor is present, contrary to the constant value obtained at high fields in the absence of the prefactor.

With the $H$ prefactor included in the fit, the $\alpha$ parameter was found to be nearly independent of temperature above 70 K with a value of $\approx 0.5$. According to our analysis of the skin depth in comparison to the dimensions of the sample surface perpendicular to the $c$ axis ($\sim$ mm), the expected value of $\alpha$ is unity according to Dyson’s theory. This discrepancy remains to be resolved.

The temperature-dependent resonant field and $g$-factor for Crystal #3 showed evidence for an unknown type of second-order phase transition at 65 K, whereas the data for Crystal #2 suggested possible transitions at 90 K and 120 K. The additional ESR signal in Crystal #3 that appears between 1 and 2 kOe in Fig. 3(b) at $T \lesssim 70$ K is likely associated with this phase transition at $\approx 65$ K rather than with paramagnetic impurities. Additional experiments are required to determine whether these features are associated with phase transitions.

The Lorentzian resonance half-width $\Delta H$ increases monotonically, but nonlinearly, from 70 K to 180 K with an average slope of 8.5 and 6.6 Oe/K for Crystals #2 and #3, respectively. These values are in the range found for similar ThCr$_2$Si$_2$-structure EuM$_2$X$_2$ compounds, where $M = \text{Fe and/or mixtures with other transition metals and } X = \text{As and/or mixtures with P.}$ We obtained microscopic information on the enhancement of the Eu moment that was reported from magnetic susceptibility measurements [2], which was the main goal of the present work. The $g$-factor obtained from the internal resonant field is found to vary nonmonotonically with temperature. For example, for Crystal #3 we find a minimum $g = 2.09$ at $\approx 100$ K and maximum values of $\approx 2.23$ at our lower and upper temperature limits of 50 K and 180 K, respectively. That the $g$-factor may be temperature dependent in the paramagnetic state was previously inferred in Ref. [2]. The temperature-averaged $g$ values for the two crystals are 2.10 and 2.15, respectively. This 5–7.5% enhancement above $g = 2$ is consistent with the 7% enhancement of the effective moment of the Eu spins found [2] from magnetic susceptibility measurements in the paramagnetic state. The $ab$ initio electronic structure calculations [2] indicated that the enhancement of the Eu moments originates from ferromagnetic polarization of the Co 3$d$ conduction electrons by the Eu spins. The present work suggests that this electron polarization is physically located around the Eu spins, and hence that the polarized $d$ electrons form a composite enhanced spin with each Eu spin. On the other hand, the saturation moments at $H = 140$ kOe obtained for crystals from the same growth batches #2 and #3 were $\mu_{\text{sat}} = 7.04$ and 7.56 $\mu_B$/Eu, respectively, where the first value is hardly enhanced and the second one is enhanced above the value expected for $g = 2$ by 8.0% [2]. The reason there was little enhancement of $\mu_{\text{sat}}$ in the crystal from batch #2 whereas the effective moment of this crystal was enhanced by 8.6% according to Tables III and IV in Ref. [2] is unclear.

A similar but opposite dichotomy to that seen for EuCo$_{2-y}$As$_2$ growth batch #2 was observed for Gd metal containing Gd$^{+3}$ spins-7/2. Whereas the low-$T$ saturation moment is enhanced from 7.00 to 7.55 $\mu_B$/Gd, the effective moment obtained from the magnetic susceptibility in the paramagnetic state above the ferromagnetic Curie temperature $T_C = 294$ K [16] is 7.98(5) $\mu_B$/Gd [17], which is the same within the errors as predicted for $g = 2$. In two separate studies, ESR measurements of Gd$^{+3}$ ions in Gd metal in the paramagnetic state also yielded unenhanced $g$-factor values of 1.95(3) and 1.97, respectively [18, 19]. It is peculiar that the low-$T$, high-$H$ saturation moment of the Gd$^{+3}$ ions in Gd metal is strongly enhanced whereas the paramagnetic effective moment and $g$-factor of the Gd$^{+3}$ ions at temperatures above $T_C$ are not.

Appendix: ESR Power Absorption by Local Magnetic Moments in Metals

When the $H$ prefactor in $\chi'_0(H)$ in Eq. (26) is omitted when fitting CW ESR data for local magnetic moments in metals by Dysonian-type lineshapes, irrespective of the values of $\alpha$, $H_{\text{res}}$ and $\Delta H$ the integral of the absorbed power over all $H$ at a given temperature $T$ is

$$\int_0^\infty P(H, T)dH \propto \chi_0(T) \quad (H \text{ prefactor absent). \hspace{1cm} (A.1)}$$

If this were valid one could compare the $T$ dependence of the integral with $\chi_0(T)$ measured with a dc magnetometer. For $\Delta H/H_{\text{res}} \ll 1$ this procedure is indeed accurate and useful if the upper limit of integration is limited to a few times the resonance field. However, we found that major differences between the two measures of $\chi_0(T)$ can occur when broad lines with $\Delta H/H_{\text{res}} \sim 1$ are observed such as in our ESR study of EuCo$_{2-y}$As$_2$ described in the main text.

Before further discussing the absorbed microwave power, we first plot the ESR power absorption spectrum itself versus reduced field $H/H_{\text{res}}$ because the shapes of such spectra for $\alpha > 0$ are perhaps unexpected. Shown in Fig. 7(a) is the conventional Lorentzian lineshape for
FIG. 7: ESR power absorption spectra with reduced Lorentzian half-width $\Delta H / H_{\text{res}} = 0.1$ versus the reduced applied field $H / H_{\text{res}}$ for $\alpha$ values of (a) 0 and (b) 1, both without and with the $H$ prefactor in Eq. (26) in the main text.

a relatively narrow spectrum with $\Delta H / H_{\text{res}} = 0.1$ with and without the $H$ prefactor in Eq. (26), where the presence or absence of the $H$ prefactor is seen to make a relatively small difference. On the other hand, when $\alpha$ takes its maximum value $\alpha = 1$, the lineshapes with and without the $H$ prefactor are significantly different as shown in Fig. 7(b). For either case with applied fields $H / H_{\text{res}} \lesssim 1$ and $\alpha = 1$, the ESR power absorbed is negative, which means that the sample is transferring energy from the temperature reservoir with which it is in thermal contact to the source of the microwave power.

Shown in Figs. 8(a) and 8(b) are data as in Fig. 7 except that the $H$ prefactor is much more pronounced for both $\alpha = 0$ and $\alpha = 1$.

The field-dependent integral of the absorbed power at a given temperature $T$ versus $H$ at constant $\omega$ is defined as

$$P_{\text{int}}(H) \propto \int_0^H \chi''(H') dH'.$$

(A.2)

If the $H$ prefactor in Eq. (26) is not present one obtains

$$\int_0^\infty \chi''(H') dH' = \pi \chi_0 \quad (\text{no } H \text{ prefactor}).$$

(A.3)

On the other hand when the $H$ prefactor is included as required by theory one has

$$\int_0^\infty \chi''_D(H') dH' = \infty \quad (\text{with } H \text{ prefactor}).$$

(A.4)

This (logarithmic) divergence occurs because for large $H$, the $H$ prefactor is much more pronounced for both $\alpha = 0$ and $\alpha = 1$. The field-dependent integral of the absorbed power at a given temperature $T$ versus $H$ at constant $\omega$ is defined as

$$P_{\text{int}}(H) \propto \int_0^H \chi''(H') dH'.$$

(A.2)

If the $H$ prefactor in Eq. (26) is not present one obtains

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(A.3)

On the other hand when the $H$ prefactor is included as required by theory one has

$$\int_0^\infty \chi''_D(H') dH' = \infty \quad (\text{with } H \text{ prefactor}).$$

(A.4)
FIG. 9: Field-integrated ESR power absorption spectra obtained from Eq. (A.2) with reduced Lorentzian half-width $\Delta H/H_{\text{res}} = 0.1$ versus the reduced applied field $H/H_{\text{res}}$ for $\alpha$ values of (a) 0 and (b) 1. If the $H$ prefactor in Eq. (26) is omitted, the integrated power is $\lim_{H \to \infty} P_{\text{int}}(H) = \pi \chi_{0}$ for $\alpha = 0$ and 1, whereas with the prefactor present one obtains $\lim_{H \to \infty} P_{\text{int}}(H) = \infty$ for either $\alpha$ value.

\[ \chi''_{D}(H) \text{ becomes} \]
\[ \chi''_{D}(H/H_{\text{res}} \gg 1) = \frac{2(\alpha + \Delta H/H_{\text{res}})}{H/H_{\text{res}}}. \]  

(A.5)

Plots of $P_{\text{int}}(H/H_{\text{res}})$ described by Eq. (A.2) with $\Delta H/H_{\text{res}} = 0.1$ for $\alpha = 0$ and 1 are shown in Fig. 9. One sees in Fig. 9(a) for $\alpha = 0$ that there is only a minor difference between the curves calculated with and without the $H$ prefactor in Eq. (26) for $H/H_{\text{res}} \leq 4$. However, for $\alpha = 1$ a major difference is seen in Fig. 9(b) between the two curves. When the linewidth increases to $\Delta H/H_{\text{res}} = 0.5$ as shown in in Fig. 10, the discrepancies between the integrated power absorption with and with-

FIG. 10: Same as Fig. 9 except that $\Delta H/H_{\text{res}} = 0.5$.

out the $H$ prefactor are increased for both $\alpha = 0$ and $\alpha = 1$ compared to the corresponding $\Delta H/H_{\text{res}} = 0.1$ data in Fig. 9.

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